Single-molecule unzippering experiments on DNA and Peyrard-Bishop-Dauxois model

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In this paper, we rely on a nonlinear Peyrard-Bishop-Dauxois (PBD) model. This mechanical model explains DNA dynamics assuming only transversal oscillations of nucleotides. The potential energy for the hydrogen bonds, connecting *AT* or *CG* base pairs, is modeled by a Morse potential. This potential is characterized by the depth D and the inverse width a of the Morse potential well. We discuss one type of single molecule manipulation experiments, which we call unzippering experiments. It is explained that the highest values of two essential parameters of the PBD model, the parameters D and a, can be determined according to the results of those experiments. This statement is supported by theoretical calculations. We show that the inverse width of the Morse potential well a has been overestimated so far. The smallest value for this parameter is determined according to the PBD model, which means that a rather narrow interval can be assumed. Also, we give an idea how to determine the optimal value of the parameter a.

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I. INTRODUCTION

It is well known that the *B*-form DNA is a right-handed double helix, which consists of two complementary strands. The helix has a diameter of about 24 Å and a helical pitch of 10.4 base pair (bp) per turn, while the vertical spacing of the basis is about 3.4 Å [1].

There have been a lot of attempts to describe DNA dynamics with appropriate models. A hierarchy of the most important models for the nonlinear DNA dynamics was presented by Yakushevich [2]. Most of them are mechanical models, like the one that we use in this paper.

The Peyrard-Bishop (PB) model was proposed in Ref. [3]. This model does not take helicoidal structure into consideration, while its improved version, which we call the Peyrard-Bishop-Dauxois (PBD) model, does [4,5].

In the past few years, a couple of single-molecule micromanipulation techniques have been used to study the structure of individual biopolymers such as DNA, RNA, and proteins. The aim of this paper is to find a relationship between the PBD model and the micromanipulation experiments. As will be seen later, those experiments could be used to determine the values, or at least the possible intervals, of some parameters used in the theoretical model.

In what follows, we outline the main features of the PBD model (Sec. II). We do not go through rather tedious mathematical derivations as this can be found in cited references.

Then, in Sec. III, we explain and discuss a couple of single-DNA micromanipulation experiments where the force between the DNA strands was measured. We call them unzippering experiments. According to those results, one can estimate the possible values of the two basic parameters of the PBD model. In fact, we estimate their highest values according to the experimental results. This is further corroborated by some theoretical estimations (Sec. IV). Also, we determine the smallest possible values of those parameters according to the theory. Finally, we suggest a procedure, based on the PBD model, which might bring about the optimal value of the parameters. This might be the most intriguing part of the paper.

It is important to keep in mind that the two kinds of estimates are on different footings. In Sec. III, we carried out the estimations according to the unzippering experiments only and no theoretical model was assumed.

In Sec. IV, however, estimations were carried out according to the PBD model, explained in Sec. II. The fact that the estimated values obtained according to those different methods are very close certainly yields to a conclusion that the PBD model is the appropriate one.

We close this paper with the summary and concluding remarks.

II. PEYRARD-BISHOP-DAUXOIS MODEL

According to both, the PB and the PBD models, one can assume a common mass m for all the nucleotides as well as the same coupling constant k along each strand. This simplification means that the DNA chain is treated as a perfectly homogeneous periodic structure.

The helicoidal structure of the DNA chain can be taken into consideration assuming that neighboring nucleotides from different strands are close enough and may interact. The nucleotide at the site n of one strand interacts with both the $(n+h)$th and $(n-h)$th nucleotides of the other strand [4,5]. As if the helix has a helical pitch of about 10 bps per turn, as was stated above, we assume $h=5$.

According to the PBD model, only transversal motions are taken into consideration and displacements of the nucleotides at the site n from their equilibrium positions are u_n and v_n for the two strands. The strands are coupled to each other through hydrogen bonds, which are supposed to be responsible for the transverse displacements of the nucleotides. The potential energy for the hydrogen bonds connecting *AT* or *CG* base pairs is modeled by a Morse potential

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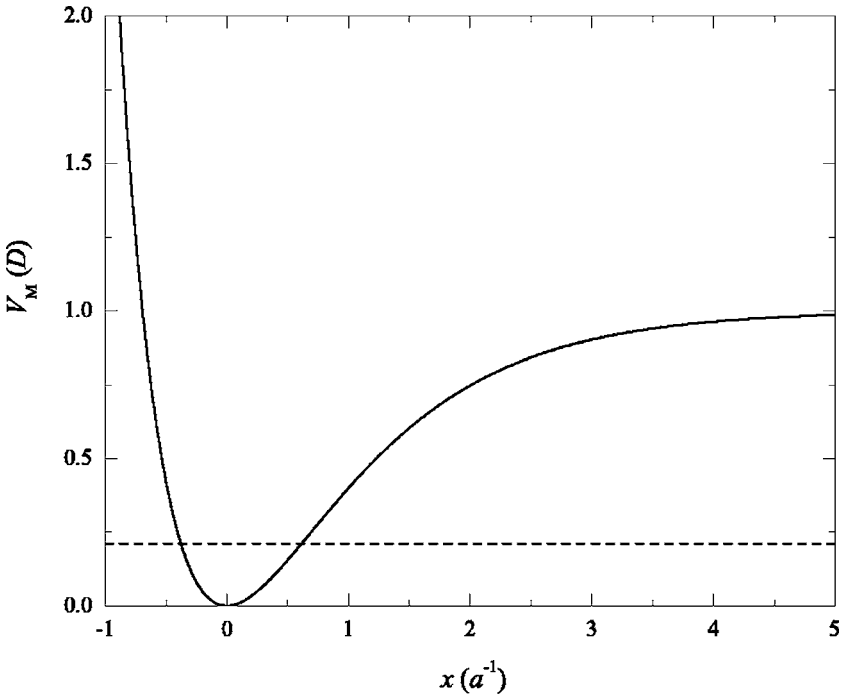

FIG. 1. Morse potential.

$$V_M(x) = D[e^{-ax} - 1]^2, \tag{1}$$

where D and a are the depth and the inverse width of the Morse potential well, respectively. To be more precise, the width of the potential is $1/a$ at $V_M \approx D/5$. This is shown in Fig. 1.

In fact, the Morse potential represents not only the hydrogen bonds, but the repulsive interactions of the phosphate, and the surrounding solvent action [4,5].

The Hamiltonian for the DNA chain is [4,5]

$$H = \sum \left\{ \frac{m}{2}(\dot{u}_n^2 + \dot{v}_n^2) + \frac{k}{2}[(u_n - u_{n-1})^2 + (v_n - v_{n-1})^2] + \frac{K}{2}[(u_n - v_{n+h})^2 + (u_n - v_{n-h})^2] + D[e^{-a(u_n - v_n)} - 1]^2 \right\}, \tag{2}$$

where k (K) is the harmonic constant of the longitudinal (helicoidal) spring.

It is more convenient to describe the motion of two strands by making a transformation to the center-of-mass coordinates representing the in-phase and out-of-phase transversal motions, namely,

$$x_n = (u_n + v_n)/\sqrt{2}, \quad y_n = (u_n - v_n)/\sqrt{2}. \tag{3}$$

The dynamical equations, derived from the Hamiltonian (2), are

$$m\ddot{x}_n = k(x_{n+1} + x_{n-1} - 2x_n) + K(x_{n+h} + x_{n-h} - 2x_n), \tag{4}$$

$$m\ddot{y}_n = k(y_{n+1} + y_{n-1} - 2y_n) - K(y_{n+h} + y_{n-h} + 2y_n) + 2\sqrt{2}aD(e^{-a\sqrt{2}y_n} - 1)e^{-a\sqrt{2}y_n}. \tag{5}$$

The first of those decoupled equations describes usual linear waves (phonons) while the second one describes nonlinear waves (breathers). Hence, we restrict our attention on the second nonlinear equation.

To solve Eq. (5), a long and tedious procedure should be performed [4–7]. All the tedious derivations and important explanations can be found in Ref. [8].

First, we assume small oscillations as

$$y = \varepsilon\Phi \quad (\varepsilon \ll 1), \tag{6}$$

which means that the nucleotides oscillate around the bottom of the Morse potential well, given in Eq. (1). However, those oscillations are still large enough to be enharmonic. Then, we use the semi-discrete approximation [8] and expect the solution to be a modulated solitonic wave [4–8]

$$\Phi_n(t) = F_1(\varepsilon nl, \varepsilon t)e^{i\theta_n} + \varepsilon[F_0(\varepsilon nl, \varepsilon t) + F_2(\varepsilon nl, \varepsilon t)e^{i2\theta_n}] + cc + O(\varepsilon^2), \tag{7}$$

$$\theta_n = nql - \omega t, \tag{8}$$

where l is the distance between neighboring nucleotides of the same strand, $\omega \equiv \omega_0$ is the optical frequency of the linear approximation, and $q = 2\pi/\lambda$ is the wave number of a carrier wave. For the most favorable mode, that is the most probable mode, we suggested [7–9] the one for which $ql = \pi/h$. The functions F_0 and F_2 can be expressed through the function F_1 [4–8], which is a solution of the nonlinear Schrödinger equation

$$iF_{1\tau} + PF_{1SS} + Q|F_1|^2F_1 = 0, \tag{9}$$

where τ and S are time and space coordinates [4–8], while the dispersion coefficient P and the coefficient of nonlinearity Q are given by

$$P = \frac{1}{2\omega}\left\{\frac{l^2}{m}[k\cos(ql) - Kh^2\cos(qhl)] - V_g^2\right\} \quad (10)$$

and

$$Q = -\frac{\omega_g^2}{2\omega}[2\alpha(\mu+\delta) + 3\beta]. \quad (11)$$

The optical frequency can be obtained from a dispersion relation [4–8]

$$\omega^2 = \omega_g^2 + \frac{2k}{m}[1-\cos(ql)] + \frac{2K}{m}[1+\cos(qhl)] \quad (12)$$

while V_g is a group velocity defined as

$$V_g \equiv \frac{d\omega}{dq} = \frac{l}{m\omega}[k\sin(ql) - Kh\sin(qhl)]. \quad (13)$$

The remaining parameters are [4–8]

$$\omega_g^2 = \frac{4a^2D}{m}, \quad (14)$$

$$\alpha = \frac{-3a}{\sqrt{2}}, \quad (15)$$

$$\beta = \frac{7a^2}{3}, \quad (16)$$

$$\mu = -2\alpha\left[1 + \frac{4K}{m\omega_g^2}\right]^{-1}, \quad (17)$$

and

$$\delta = \omega_g^2\alpha\left[4\omega^2 - \frac{2k}{m}[1-\cos(2ql)] - \frac{2K}{m}[1+\cos(2hql)] - \omega_g^2\right]^{-1}. \quad (18)$$

For $PQ>0$, Eq. (9) has an envelope soliton solution [4,5,10]. Also, as was stated previously, functions F_0 and F_2 can be expressed through the function F_1. Therefore, we can finally obtain the expression for the functions $y(t)$ and $\Phi_n(t)$, defined by Eqs. (6)–(8) as [4–8]

$$\Phi_n(t) = 2A\,\mathrm{sech}\left(\frac{\varepsilon(nl - V_e t)}{L_e}\right)\left\{\cos(\Theta nl - \Omega t) + \varepsilon A\,\mathrm{sech}\left(\frac{\varepsilon(nl - V_e t)}{L_e}\right)\times\left[\frac{\mu}{2} + \delta\cos[2(\Theta nl - \Omega t)]\right]\right\} + O(\varepsilon^2), \quad (19)$$

where

$$\Theta = q + \frac{\varepsilon u_e}{2P}, \quad (20)$$

$$V_e = V_g + \varepsilon u_e, \quad (21)$$

$$\Omega = \omega + \frac{(V_g + \varepsilon u_c)\varepsilon u_e}{2P}, \quad (22)$$

$$A = \sqrt{\frac{u_e^2 - 2u_e u_c}{2PQ}}, \quad (23)$$

$$L_e = \frac{2P}{\sqrt{u_e^2 - 2u_e u_c}}, \quad (24)$$

and u_e and u_c are the velocities of the envelope and the carrier waves, respectively.

We showed [11] that this solution exists only if $K<a^2D$. Otherwise, the parameter Q would be negative and the amplitude A would be imaginary.

The above model is illustrated by Figs. 2 and 3 for $n=300$. One can see that the function $\Phi(t)$ is a modulated solitonic wave called breather.

We chose the following set of values for parameters characterizing a traveling wave solution [4,5]

$$u_e = 10^5\ \mathrm{m/s}, \quad u_c = 0, \quad \varepsilon = 0.007. \quad (25)$$

Figures 2 and 3 were carried out for $k=24$ N/m [4,5], $K=4$ N/m and for $k=1$ N/m [12–14], $K=0.5$ N/m, respectively. We discuss those values in the next section. For both figures, we picked up $a=2$ Å^{-1} and $D=0.1$ eV [4,5] as well known values $l=3.4\times10^{-10}$ m and $m=5.1\times10^{-25}$ kg. One can find different values for parameters a and D, but we will return to this issue later.

We defined [11] a density of internal oscillations (density of carrier wave oscillations) as

$$D_0 \equiv \frac{\Lambda}{\lambda_c}, \quad (26)$$

where Λ and λ_c are the length of the envelope and the wavelength of the carrier wave, respectively. To be more precise, this is a number of the carrier wavelengths per an envelope, but we use the word density for short. From hyperbolic and cosine terms in expression (19), one can see that

$$\Lambda = \frac{2\pi L_e}{\varepsilon} \quad (27)$$

and

$$\lambda_c = \frac{2\pi}{\Theta} = \frac{2\pi}{q + \frac{\varepsilon u_e}{2P}}. \quad (28)$$

From Eqs. (24)–(28), we can easily obtain

$$D_0 = 1 + \frac{2qlP}{\varepsilon u_e l}. \quad (29)$$

The function D_0 describes the solitonic solution $\Phi(nl)$, which is Eq. (19) for a particular value of t, rather then $\Phi(t)$, given by Figs. 2 and 3 for $n=300$. Therefore, we should also define the density of internal oscillations (density of carrier wave oscillations) Γ_0 as a ratio of two periods. According to Eq. (19), this function is

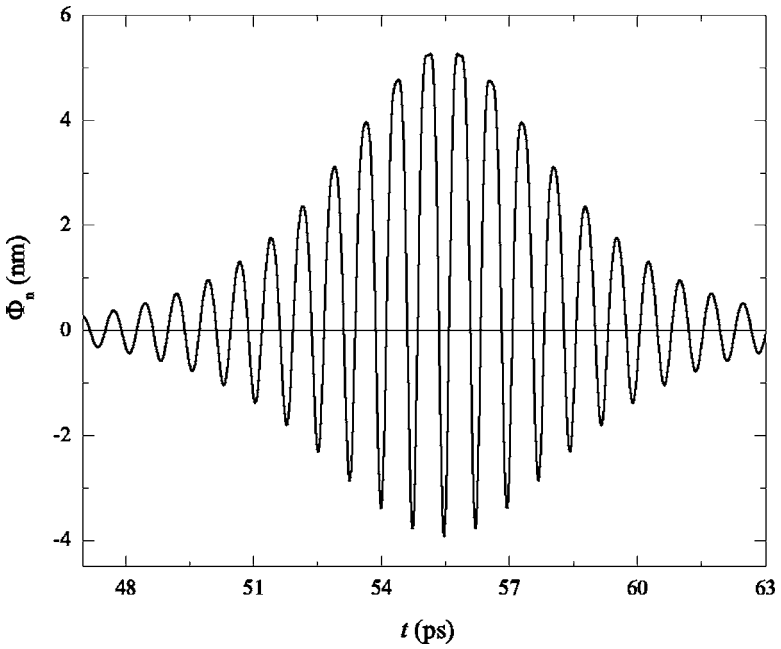

FIG. 2. Elongation of the out-of-phase motion as a function of time (k=24 N/m, K=4 N/m, D=0.1 eV, a=2 Å^{-1}).

$$\Gamma_0 = \frac{\Omega}{\varepsilon V_e / L_e}. \tag{30}$$

From Eqs. (21), (22), (24), (25), and (30) and for $V_g \gg \varepsilon u_e$, which might not be always correct, one easily obtains

$$\Gamma_0 = 1 + \frac{2\omega P}{\varepsilon u_e V_g}. \tag{31}$$

For Figs. 2 and 3, we can calculate, according to Eq. (30), Γ_0=20.6 and Γ_0=5.8, respectively.

III. EXPERIMENTS AND DISCUSSIONS

A first single DNA molecule experiment was carried out in 1992 [15]. That was a direct mechanical micromanipulation of the single DNA. From then, a few techniques have been used to determine elastic properties of the molecules and to induce structural transitions. Most of those micromanipulation experiments are stretching experiments [16–22]. Usually, the force-displacement response of the single DNA molecule was measured. Those experiments were followed by theoretical research [23–28] and computer simulations [29]. This research has also been reviewed [1,14].

However, in this work, we are interested in the two parameters characterizing the Morse potential, as was stated

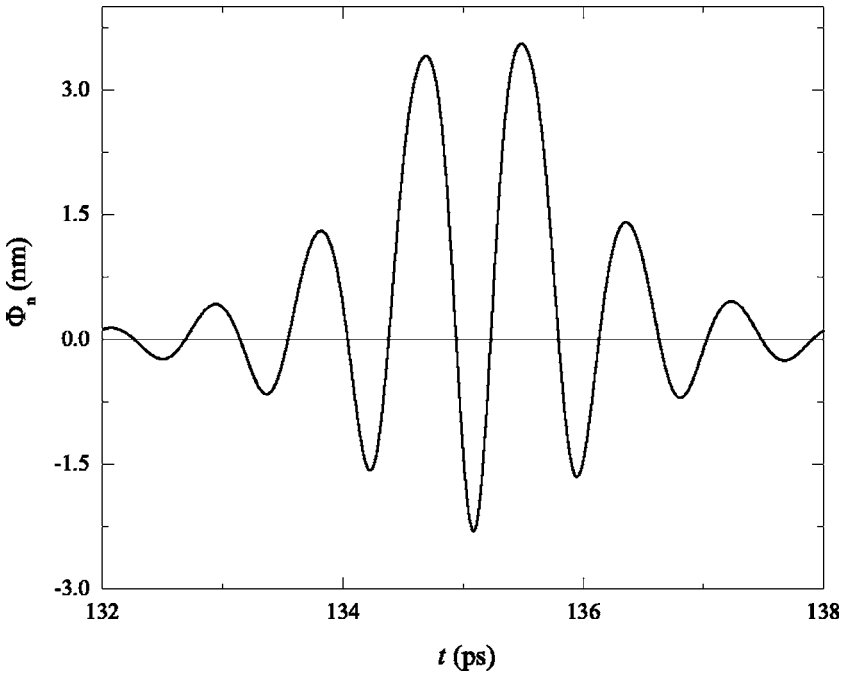

FIG. 3. Elongation of the out-of-phase motion as a function of time (k=1 N/m, K=0.5 N/m, D=0.1 eV, a=2 Å^{-1}).

TABLE I. Pairs of the Morse potential parameters: a (inverse width) and D (depth of the Morse potential well).

a (Å^{-1})	D (eV)	Reference	aD (pN)
1.4	0.19	[37]	425.6
1.8	0.33	[3]	950.4
2	0.1	[4,5]	320
4.45	0.04	[38]	284.8
4.5	0.03	[14]	216
6.3	0.15	[36]	1512
AT 4.2	0.05	[39]	336
GC 6.9	0.075	[39]	828

previously. As if this potential is responsible for the transverse force between the two strands, we discuss results of a couple of experiments where the interaction between the two DNA strands was measured. We call them unzippering experiments. The results of those experiments will be used to test the PBD model. In fact, we show that one can determine the highest value of the product aD, where a and D are the parameters explained previously. We will see that the value of the parameter a has been overestimated in probably all the papers dealing with it.

In some of those unzippering experiments [30–32], mechanical strand separation was carried out. The 3′ and 5′ extremities on one end of the molecule are pulled progressively apart, and this leads to the opening of the double helix. In Ref. [30], the opening forces in the range of 10–15 pN were reported. It was roughly estimated that the value of the force would be about 10 pN for opening a 100% *AT* sequence and about 15 pN for a 100% *GC* sequence.

It was determined [31] that the DNA molecule starts to open when the force approaches 12 pN, while, during the unzippering of the two strands, the force signal shows variations between 11–14 pN.

In Ref. [32], the progressive opening was carried out for different opening velocities. It is reported that the average value and amplitude of the force signal are almost independent of the opening velocity in the interval 20 nm/s to 800 nm/s. Also, a rapid variation of the amplitude of about 2 pN was reported. This could be explained by the fact that the opening force might be fragment dependent.

For some papers, the name "unzippering experiments" is not convenient as if the interchain interaction was carried out in a different way. Namely, the complementary segments were covalently attached to opposing surfaces [33]. For example, force versus relative surface displacement was measured between $(ACTG)_{5'}$ and $(CAGT)_{5'}$ functionalized surfaces. The highest forces for 20, 16, and 12 bps were 1520, 1110, and 830 pN, respectively. This means that the highest force was 76 pN/bp.

In a similar experiment [34], the DNA oligomer **a**=5′-*G-G-C-T-C-C-C-T-T-C-T-A-C-C-A-C-T-G-A-C-A-T-C-G-C-A-A-C-G-G*-3′ was tested against its compliment **b**=3′-*C-C-G-A-G-G-G-A-A-G-A-T-G-G-T-G-A-C-T-G-T-A-G-C-G-T-T-G-C-C*-5′ and the truncated complements **c**=3′-*A-T-G-G-T-G-A-C-T-G-T-A-G-C-G-T-T-G-C-C*-5′ and **d**=3′-*T-A-G-C-G-T-T-G-C-C*-5′. Therefore, the lengths of sequences **c** and **d** are 20 and 10 bps, while complementary oligomers **a** and **b** are 30 bps long. The unbinding forces were in the interval 20 pN to 50 pN. A close result, 54 pN, was also reported [35].

In a theoretical paper [36], the highest force was calculated to be about 275 pN. However, about 4 bps were involved in the interaction, which means that the interaction force is about 69 pN/bp.

As a conclusion, we can state that the highest reported value of the unbinding force is approximately

$$F_{em} \approx 75 \text{ pN}. \tag{32}$$

A. Estimations according to the experimental values

As was stated above, the aim of this section is to estimate the highest value of the parameter aD. It was explained that the DNA strands were coupled to each other through hydrogen bonds. The potential energy of this interaction, connecting base pairs of the different strands, is modeled by the Morse potential, given by Eq. (1) and the last term in Eq. (2). Note that the stretching of the nucleotides, belonging to the same pair, is

$$u - v = y\sqrt{2} = z \tag{33}$$

as can be seen from Eqs. (1)–(3).

A force coming from the Morse potential can be easily calculated according to Eq. (1) as a first derivative of the function V_M. The highest value of this Morse force is

$$F_{Mm} = \frac{aD}{2}. \tag{34}$$

As was stated above, different experimental values for unbinding forces have been reported so far [30–36]. Those forces are from about 10 pN/bp to approximately 75 pN/bp. This brings about a conclusion that the calculated force (34) should be less than the maximal reported experimental force F_{em}, that is

$$\frac{aD}{2} \leqslant F_{em}. \tag{35}$$

However, Eq. (35) requires further discussions. One can ask if Eq. (35) would be correct if the experimental value F_{em} were correct. In other words, we need to study the possibility that the experiments, explained in Ref. [33], are perfect and that we can assume that the value of F_{em} =75 pN/bp is exactly the highest value of the unbinding force. Suppose for a moment that the Morse force represents all the possible interactions between the nucleotides. If so, will the left and the right side of Eq. (35) be equal? The answer depends on the type of the experiments. To see this, we need to apply the following well known formulas to the DNA molecule

$$dU = TdS + dA, \tag{36a}$$

TABLE II. The smallest values of the parameter a calculated from the requirement $Q(a) \geqslant 0$ and the double values of the maximum of the Morse force $F_{Mm}=aD/2$.

Triplet	k (N/m)	K (N/m)	D (eV)	$\alpha_{\min}$ (Å^{-1})	$\alpha_{\min}D$ (pN)
	1	0.5	0.15	0.558	133.9
1	1	0.5	0.10	0.683	109.3
	1	0.5	0.05	0.966	77.3
	1	0.3	0.15	0.430	103.2
2	1	0.3	0.10	0.527	84.3
	1	0.3	0.05	0.745	59.6
	3	1	0.15	0.786	188.6
3	3	1	0.10	0.963	154.1
	3	1	0.05	1.362	108.9
	3	0.5	0.15	0.550	132.0
4	3	0.5	0.10	0.673	107.7
	3	0.5	0.05	0.952	76.2
	6	2	0.15	1.112	266.9
5	6	2	0.10	1.362	217.9
	6	2	0.05	1.926	154.1
	6	0.5	0.15	0.538	129.1
6	6	0.5	0.10	0.659	105.4
	6	0.5	0.05	0.932	74.6
	24	4.3	0.15	1.615	387.6
7	24	4.3	0.10	1.978	316.5
	24	4.3	0.05	2.800	223.8
	24	3	0.15	1.337	320.9
8	24	3	0.10	1.638	262.1
	24	3	0.05	2.316	185.3
	24	1	0.15	0.730	175.2
9	24	1	0.10	0.894	143.0
	24	1	0.05	1.264	101.1
	24	0.5	0.15	0.480	115.1
10	24	0.5	0.10	0.587	94.0
	24	0.5	0.05	0.831	66.4

$$dF = -SdT + dA, \tag{36b}$$

where U and F are internal energy and Helmholtz free energy, respectively, and dA is work done on the system. For the experiments explained in Ref. [33], one can safely assume both $S=$const and $T=$const. If we assume that the potential energy part of U is V_M and that the experimental force is the first derivative of the free energy, then both sides in Eq. (35) are equal.

The only problem might be if the Morse potential does not represent all the possible interactions. It was stated previously that the Morse potential represents not only the hydrogen bonds, but the repulsive interactions of the phosphate, and the surrounding solvent action [4,5]. If so, we can state that $aD/2$ is, if not less then, than certainly approximately equal to F_{em}, corroborating Eq. (35).

In papers, one can find various choices for the parameters a and D. Some of those values are given in Table I. In the last two rows, the values for AT and GC pairs are given.

Obviously, all the choices for a and D are unacceptable as they do not satisfy Eqs. (32) and (35). In other words, both parameters have been overestimated. The only exception may be small values for D. Namely, a small value for D, like $D=0.05$ eV, together with very small a, may satisfy Eqs. (32) and (35). For example, for $a=1$ Å^{-1} and $D=0.05$ eV, one can calculate $aD=80$ pN<150 pN. Therefore, we state that the value of the parameter a has certainly been overestimated. As if $1/a$ is the width of the Morse potential, we can conclude that DNA is "softer" then it has been assumed so far.

Finally, we want to point out that Eq. (35) is not related to any solution of Eq. (5). In other words, no physical model, describing DNA dynamics, was assumed. To be more precise, only what is common for Eq. (35) and the PBD model is the Morse potential, given by Eq. (1). However, theoretical estimations will be relied on the PBD model, which is a topic of the next section. Those estimations will be compared with the experimental values, e.g., with Eqs. (32) and (35).

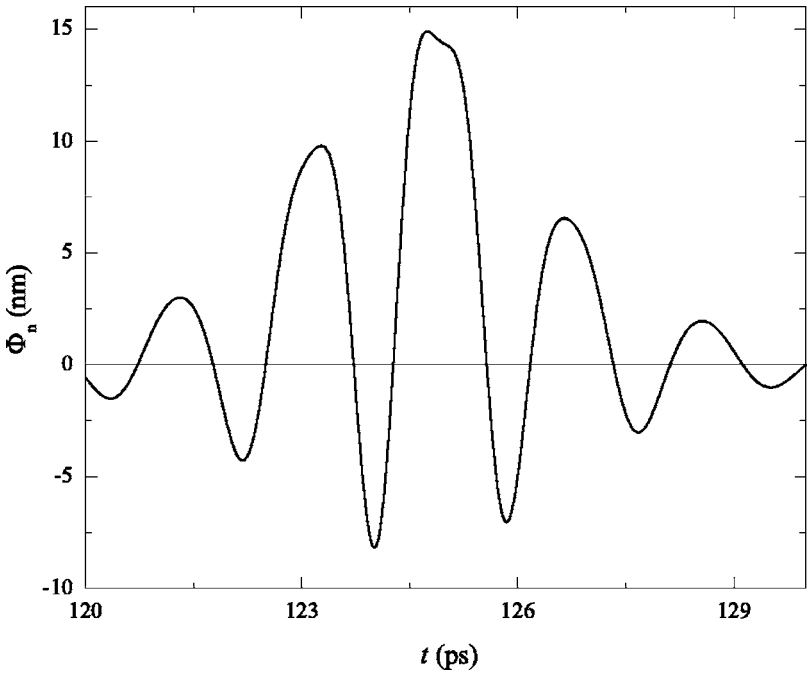

FIG. 4. Elongation of the out-of-phase motion as a function of time (k=1 N/m, K=0.5 N/m, D=0.1 eV, a=0.9 Å^{-1}).

IV. ESTIMATIONS ACCORDING TO THE PBD MODEL

In what follows, we estimate the values of the parameters a and D according to the PBD model of the DNA molecule. We show that those estimations are in good agreement with the expressions (32) and (35).

A. Estimation 1

Let us study a function

$$f(z) = (e^{-az} - 1)e^{-az}. \tag{37}$$

This is the last term in Eq. (5) where $y\sqrt{2}$ was replaced by z according to Eq. (33). Therefore, z is the stretching of the nucleotide pair. To derive Eq. (9), we performed a series expansion of the exponential terms. Of course, this is correct only for very small values for az. In other words, instead of the function (37), a new approximated one

$$F(z) = \left(1 - az + \frac{a^2z^2}{2} - \frac{a^3z^3}{6} - 1\right)\left(1 - az + \frac{a^2z^2}{2}\right)$$
$$\approx -az + \frac{3}{2}a^2z^2 - \frac{7}{6}a^3z^3 \tag{38}$$

was used. It is easy to see that there should be

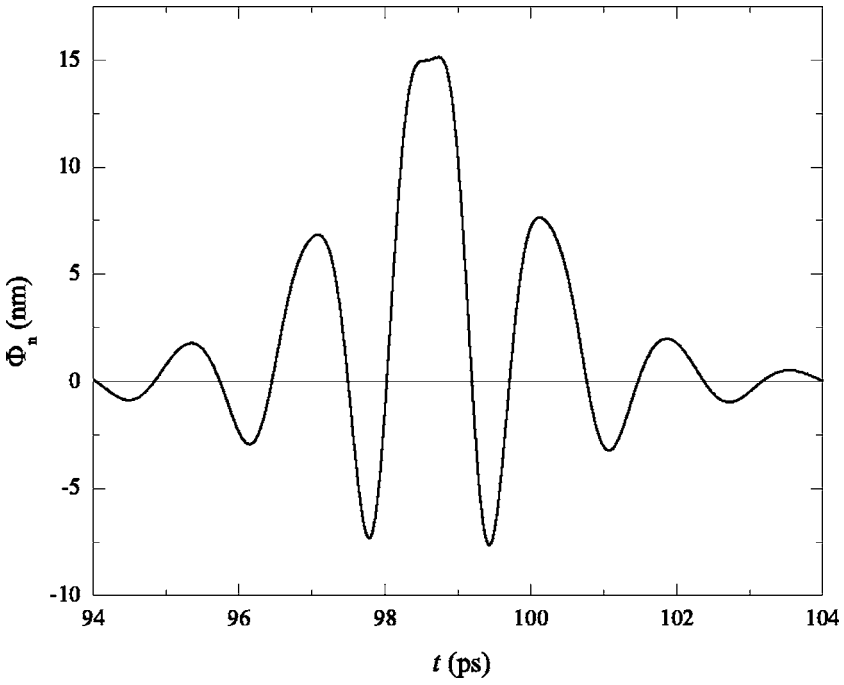

FIG. 5. Elongation of the out-of-phase motion as a function of time (k=3 N/m, K=0.5 N/m, D=0.1 eV, a=0.9 Å^{-1}).

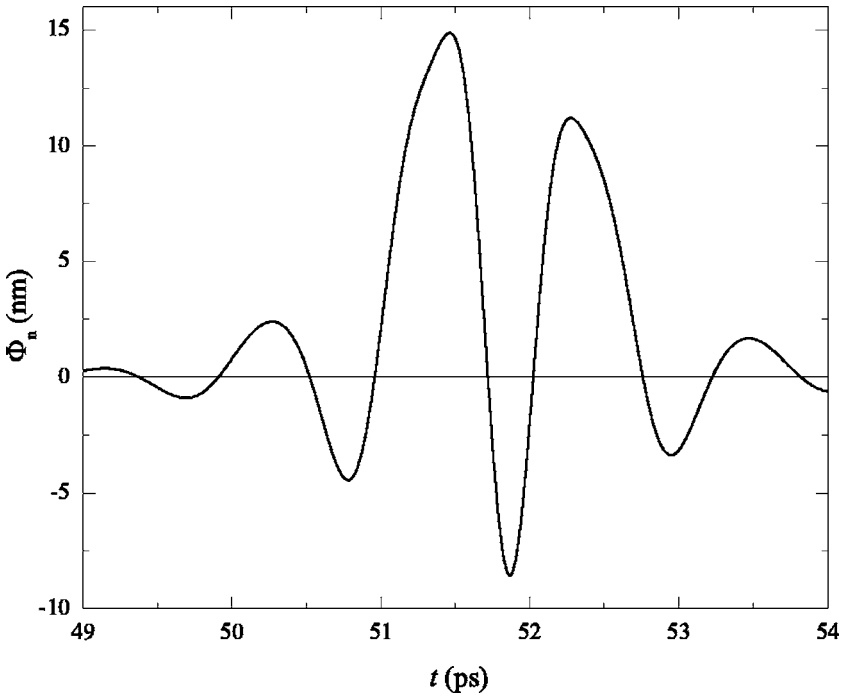

FIG. 6. Elongation of the out-of-phase motion as a function of time ($k=15$ N/m, $K=0.5$ N/m, $D=0.1$ eV, $a=0.9$ Å^{-1}).

$$ay \leqslant 0.3 \tag{39}$$

for the error not to exceed about 10%. Hence, large a is not compatible with the used theory. For example, for $a=1.5$ Å^{-1} the stretching $z=y\sqrt{2}$ should be less then 0.3 Å. This means that the amplitudes of the oscillating nucleotides, which is half of the stretching, would be smaller then 0.15 Å, which is extremely small. Note that the distance between the nucleotides of the same pair is about 3 Å. Hence, if we assume that the amplitude of the oscillating nucleotide should be higher then 0.2 Å, then there should be $a<1$ Å^{-1}.

Now, we can estimate the upper limit of the parameter D. According to Eqs. (32) and (35), one obtains

$$D \leqslant 0.09 \text{ eV} \quad \text{for } a \approx 1 \text{ Å}^{-1},$$

$$D \leqslant 0.12 \text{ eV} \quad \text{for } a \approx 0.8 \text{ Å}^{-1},$$

$$D \leqslant 0.16 \text{ eV} \quad \text{for } a \approx 0.6 \text{ Å}^{-1}.$$

B. Estimation 2

The highest values for the parameters a and D have been discussed so far. The aim of this section is to discuss the smallest value for a.

As was stated above, the solitonic solution (19) exists if $PQ>0$. One can easily check that the dispersion coefficient P is always positive. Hence, we want to determine how the nonlinear parameter Q depends on a. However, Q also depends on a few more parameters. This is why the smallest value for the parameter a, for which $Q=0$, is showed in a Table II for a couple of values of the parameters k, K, and D. In other words, the parameter Q is an increasing function of a and Table II was carried out according to the figures $Q(a)$ and the requirements $Q(a)\geqslant 0$.

We should keep in mind that $Q=0$ implies $A\to\infty$ according to Eq. (23). This means that a cannot be infinitely close to $a_{\min}$.

The optical frequency is given by Eq. (12). If we had studied phonons, we would have obtained the acoustical frequency. It was showen [40] that the optical frequency is higher than the acoustical one if

$$K < a^2 D. \tag{40}$$

Equality in Eq. (40) would imply the resonance mode [40,41]. One can easily check that Eq. (40) holds for all the triplets in Table II.

Table II shows that $a_{\min}$ depends much more on K than on the parameter k. We can conclude that large values for K is not a good choice. Namely, no triplet for $K\geqslant 1$ N/m is convenient as if either a or aD is too large. Also, $D=0.05$ eV is probably too small. On the other hand, we cannot exclude any value of the parameter k.

In Figs. 4–6, we show the breather $\Phi_n(t)$ for the inverse width of the Morse potential a belonging to the above suggested interval. All of them were carried out for $a=0.9$ Å^{-1}, $D=0.1$ eV, and $K=0.5$ N/m. For the parameter k, we picked up 1 N/m, 3 N/m, and 15 N/m. We do not have to worry about very large amplitudes because, as was explained above, the amplitude depends on unknown parameters ε, u_e, and u_c.

Let us compare Figs. 4–6 with Figs. 2 and 3, carried out for $a=2$ Å^{-1}. Obviously, the first of them, that is Fig. 2, differs from others in having much higher density of internal oscillations ($D_0=20.6$), while D_0 for Figs. 3–6 is 5.8, 5.4, 4.8, and 3.5, respectively. From the point of view of engineering modulation, the first choice, Fig. 2, would be the best. However, such a conclusion may be wrong here. We should keep in mind that relatively big particles, nucleotides, oscillate in DNA, not electromagnetic field vector. It will be

TABLE III. The optimal values of the Morse parameter a obtained from the requirement $D_0(a_0)=\Gamma_0(a_0)$.

Triplet	K (N/m)	K (N/m)	D (eV)	a_0 (Å^{-1})
	1	0.3	0.15	0.468
2	1	0.3	0.10	0.573
	1	0.3	0.05	0.810
	3	0.5	0.15	0.473
4	3	0.5	0.10	0.579
	3	0.5	0.05	0.819
	6	0.5	0.15	0.532
6	6	0.5	0.10	0.652
	6	0.5	0.05	0.922
	24	0.5	0.15	0.712
10	24	0.5	0.10	0.872
	24	0.5	0.05	1.233

suggested, in the next section, that we should be inclined towards the optimal D_0, rather than the highest one.

C. Estimation 3

In the previous sections, we studied the possible interval for the parameter a. The upper limit was estimated according to both the experimental data and the PBD model, while the lower limit was discussed according to the theory (Sec. IV B). In this section, we try to study physical conditions that might bring about the optimal value for a. Unfortunately, we will not be able to determine the exact value of the parameter a because the arbitrary parameters ε, u_e, and u_c are involved in the issue.

The solitonic wave $\Phi_n(t)$ is a modulated signal and we defined the densities of the internal oscillations D_0 and Γ_0 by Eqs. (29) and (30). We suggest that the optimal value a_0 is the one which allows those two functions to be equal, that is

$$D_0(a_0) = \Gamma_0(a_0). \tag{41}$$

One can see, from Eqs. (19), (26)–(28), and (30), that the requirement (41) represents the mode when phase velocities of both the envelope and the carrier signal are equal, i.e.

$$\frac{\Omega}{\Theta} = V_e. \tag{42}$$

This is sort of a coherent mode, meaning that the wave is unchanged in time. In other words, the pattern shown in any of the Figs. 2–6 is the same at any position n of the DNA molecule.

In Table III, we show the values of a_0 for the four triplets existing in Table II. Those triplets correspond to the smallest values of K. One can see that a_0 is higher then $a_{\min}$ for the triplets 2 and 10, for triplet 6 they are almost equal, while for triplet 4, a_0 is smaller then $a_{\min}$. However, we should not worry about this inconvenience because a_0 depends on still arbitrary parameters εu_e and εu_c, as was stated previously. This means that, for a very small increase of εu_e, the parameter a_0 becomes higher then $a_{\min}$, that is between $a_{\min}$ and $a_{\max}$. The relationship between εu_e and a_0, determined according to Eq.(41), is shown in Fig. 7. The figure was carried out for $u_c=0$ [4,5]. One can see that a_0 is an increasing function with respect to εu_e for all accepted values of the parameters k and K. It might be interested to point out that the increase is the sharpest for the smallest k (line d). Therefore, very small increase of εu_e ensures a_0 to be in an expected interval, that is between $a_{\min}$ and $a_{\max}$.

A patient reader may ask how a_0 depends on εu_c. The parameter a_0 is the decreasing function on εu_c. According to Eq. (41), we can study how a_0 depends on both εu_e and εu_c. For example, one can check that, for $\varepsilon u_e=900$ m/s and

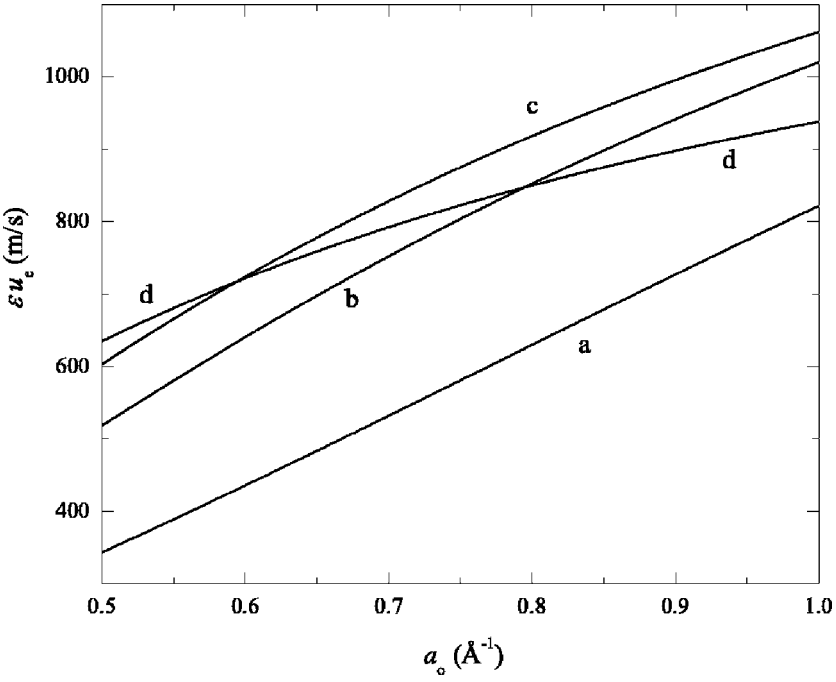

FIG. 7. Velocity εu_e as a function of the inverse width (a: k =24 N/m, K=0.5 N/m, b: k =6 N/m, K=0.5 N/m, c: k =3 N/m, K=0.5 N/m, d: k =1 N/m, K=0.3 N/m).

$\varepsilon u_c = 0.4 \varepsilon u_e$, the values of a_0 for $D=0.1$ eV are 0.699 Å^{-1}, 0.686 Å^{-1}, 0.756 Å^{-1}, and 0.973 Å^{-1} for triplets 2, 4, 6, and 10, respectively.

The previously mentioned value for g, that is $g=0.27$, comes from $D_0(g)=\Gamma_0(g)$.

V. CONCLUDING REMARKS

The main aim of this paper was to study the highest possible values of the parameters a and D, existing in the PBD model. This was determined according to the experimental data. We wanted to show that the commonly used values of those parameters have been overestimated. Our statement about $a_{\max}$ was corroborated with the simple theoretical calculations. Also, the smallest possible values were calculated according to the requirement $Q(a) \geqslant 0$. Hence, we suggested an interval for a with rather precise lower limit and the estimated, according to the experimental data, upper limit.

In addition, we gave an argument that the most favorable, that is biologically the best, mode might be the one when the phase velocities of the envelope and the carrier wave are equal. This corresponds to the requirement (41) and we called this as the coherent mode. We showed that the values of the parameter a, corresponding to the coherent mode, may be in the above mentioned interval for reasonable values of still arbitrary parameters εu_e and εu_c.

Finally, we want to express an idea how to determine the possible values for the parameters εu_e and εu_c. Namely, the requirement that a be between $a_{\min}$ and $a_{\max}$ might bring about the accepted interval for the parameters εu_e and εu_c. However, this requires a further research and is not a topic of this paper.

In this paper, the effects of viscous environment are disregarded since in Ref. [9] one approximative approach was already introduced. Nevertheless, more precise derivations of DNA dynamics in the presence of viscosity have been carried out and will be submitted soon.

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[1] T. R. Strick, M. N. Dessinges, G. Charvin, N. H. Dekker, J. F. Allemand, D. Bensimon, and V. Croquette, Rep. Prog. Phys. **66**, 1 (2003).
[2] L. V. Yakushevich, *Nonlinear Physics of DNA* (Wiley, Chichester, 1998).
[3] M. Peyrard and A. R. Bishop, Phys. Rev. Lett. **62**, 2755 (1989).
[4] T. Dauxois, Phys. Lett. A **159**, 390 (1991).
[5] T. Dauxois and M. Peyrard, in *Nonlinear Coherent Structures in Physics and Biology*, Vol. 393, edited by M. Remoissenet and M. Peyrard (Proceedings, Dijon, France, 1991), pp. 79–86.
[6] M. Remoissenet, Phys. Rev. B **33**, 2386 (1986).
[7] S. Zdravković, J. A. Tuszyński, and M. V. Satarić, J. Comput. Theor. Nanosci. **2**, 1 (2005).
[8] S. Zdravković, in *Finely Dispersed Particles: Micro-, Nano-, and Atto-Engineering*, edited by A. M. Spasic and J. P. Hsu, 130 Surfactant Science Series, (Dekker/CRC Press/Taylor & Francis Group, Boca Raton, Florida, 2005), pp. 779–811.
[9] S. Zdravković and M. V. Satarić, Phys. Scr. **64**, 612 (2001).
[10] A. C. Scot, F. Y. F. Chu, and D. W. McLaughlin, Proc. IEEE **61**, 1443 (1973).
[11] S. Zdravković, M. V. Satarić, and J. A. Tuszyński, J. Comput. Theor. Nanosci. **1**, 171 (2004).
[12] T. Dauxois and M. Peyrard, Phys. Rev. E **51**, 4027 (1995).
[13] T. Lipniacki, Phys. Rev. E **64**, 051919 (2001).
[14] M. Peyrard, Nonlinearity **17**, R1 (2004).
[15] S. B. Smith, L. Finzi, and C. Bustamante, Science **258**, 1122 (1992).
[16] D. Bensimon, A. J. Simon, V. Croquette, and A. Bensimon, Phys. Rev. Lett. **74**, 4754 (1995).
[17] P. Cluzel, A. Lebrun, C. Heller, R. Lavery, J-L. Viovy, D. Chatenay, and F. Caron, Science **271**, 792 (1996).
[18] S. B. Smith, Y. Cui, and C. Bustamante, Science **271**, 795 (1996).
[19] J. F. Allemand, D. Bensimon, R. Lavery, and V. Croquette, Proc. Natl. Acad. Sci. U.S.A. **95**, 14152 (1998).
[20] J. F. Leger, G. Romano, A. Sarkar, J. Robert, L. Bourdieu, D. Chatenay, and J. F. Marko, Phys. Rev. Lett. **83**, 1066 (1999).
[21] H. Clausen-Schaumann, M. Rief, C. Tolksdorf, and H. E. Gaub, Biophys. J. **78**, 1997 (2000).
[22] M. C. Williams, K. Pant, I. Rouzina, and R. L. Karpel, Spectroscopy (Amsterdam) **18**, 203 (2004).
[23] I. Rouzina and V. A. Bloomfield, Biophys. J. **80**, 882 (2001).
[24] M. C. Williams, I. Rouzina, and V. A. Bloomfield, Acc. Chem. Res. **35**, 159 (2002).
[25] W. M. Wartell and A. S. Benight, Phys. Rep. **126**, 67 (1985).
[26] S. M. Bhattacharjee, J. Phys. A **33**, L423 (2000).
[27] D. K. Lubensky and D. R. Nelson, Phys. Rev. Lett. **85**, 1572 (2000).
[28] S. Cocco, R. Monasson, and J. F. Marko, Proc. Natl. Acad. Sci. U.S.A. **98**, 8608 (2001).
[29] A. Lebrun and R. Lavery, Nucleic Acids Res. **24**, 2260 (1996).
[30] B. Essevaz-Roulet, U. Bockelmann, and F. Heslot, Proc. Natl. Acad. Sci. U.S.A. **94**, 11935 (1997).
[31] U. Bockelmann, B. Essevaz-Roulet, and F. Heslot, Phys. Rev. Lett. **79**, 4489 (1997).
[32] U. Bockelmann, B. Essevaz-Roulet, and F. Heslot, Phys. Rev. E **58**, 2386 (1998).
[33] G. U. Lee, L. A. Chrisey, and R. J. Colton, Science **266**, 771 (1994).
[34] T. Strunz, K. Oroszlan, R. Schäfer, and H.-J. Güntherodt, Proc. Natl. Acad. Sci. U.S.A. **96**, 11277 (1999).
[35] T. Boland and B. D. Ratner, Proc. Natl. Acad. Sci. U.S.A. **92**, 5297 (1995).

[36] S. Cocco, R. Monasson, and J. F. Marko, Phys. Rev. E **65**, 041907 (2002).
[37] Y. Gao, K. V. Devi-Prasad, and E. W. Prohofsky, J. Chem. Phys. **80**, 6291 (1984).
[38] T. Dauxois, M. Peyrard, and A. R. Bishop, Phys. Rev. E **47**, 684 (1993).
[39] A. Campa and A. Giansanti, Phys. Rev. E **58**, 3585 (1998).
[40] S. Zdravković and M. V. Satarić, Chin. Phys. Lett. **22**, 850 (2005).
[41] S. Zdravković and M. V. Satarić, Phys. Rev. E (to be published).