Topological interaction between polymer chains

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The problem of topological restrictions on a system consisting of two closed polymer chains is considered. The condition of conservation of the topological state of the system (absence of linkage, or linkage of a definite type) leads to entropy-type interaction between the chains. Such topological interactions are calculated by the Monte Carlo method. The linkage algebraic invariant (the Alexander polynomials), which offers some important advantages over the integral "potential" between unlinked polymer molecules and molecules forming a simplest type of linkage, is employed. It is found that the interaction "potentials" between unlinked polymer molecules and molecules forming a simplest type of linkage can be described by simple formulas. The interaction "potential" for unlinked molecules is used to calculate the second virial coefficient for a gas or solution of closed polymer chains. The calculations show that if topological restrictions are taken into account the polymer coils are practically impenetrable to one another. Some applications of the results are discussed.

In the construction of a consistent statistical theory of polymer chains, essential difficulties arise due to the need of taking into account various types of topological restrictions. The simplest problem of this type is that of the number of states of a chain that goes around a fixed curve in space a specified number of times (or a point on a plane). An exhaustive analytic solution for this problem was obtained in[1-5].

The next and much more complicated problem is that of taking into account the topological restrictions in the statistical mechanics of closed polymer chains. This problem was considered by Edwards[6], who used the technique of integration in functional space (see also[7-9]). The realization of this approach meets with serious difficulties. First, it is necessary to have an integral topological invariant of the knot (when a single chain is considered) or of the linkage (with two or more chains are considered). At the same time, the known Gaussian integral invariant for linkages has serious shortcomings (see below, and the invariant proposed by Edwards for the knot[7] is generally incorrect (see[10]). Second, even if the Gaussian invariant is used for linkages, the calculation of the statistical integral involves tremendous technical difficulties which have not yet been overcome.

It is therefore of interest to attempt a numerical solution of the problems of this type by using the Monte Carlo method. Numerical examples are particularly attractive in this case because they make it possible to use for the knots and linkages the invariants developed in algebraic topology. With the aid of such an approach we have obtained in an earlier paper[10] estimates of the probability of production of knots of various types for isolated chains of different length. In this paper we use an analogous approach for the study of topological interactions between different polymeric chains.

ALGEBRAIC INVARIANT OF LINKAGE

The algorithm for calculating the invariant of a linkage can be constructed in analogy with the process of calculating the invariant of a knot (the Alexander polynomial), as proposed in[10]. Just as in[10], we present this algorithm in final form, without going into the mathematical details, which can be found in[11] (see also[12]).

We introduce first a number of definitions. Two linkages are equivalent if they can be converted one from the other by continuous deformation of the contours without intersections between them or without self-intersections. Each linkage can be specified by its projection on a certain plane, which we designate by (x, y), and the projection is chosen such that the linkage is situated in a regular position. Then the points of the plane (x, y), which contain projections of more than one linkage point, will always be double, and each double point corresponds to an intersection. Just as in the case of the projection of a knot, that point whose coordinate z is smaller is called underpassing, and the other is called respectively overpassing. For linkages, the underpassing and overpassing points of a given intersection can be projections of points belonging to different contours. We confine ourselves to linkages consisting of two contours. We choose on each of these contours (S and T) one arbitrary point (0), which is not a double point, and specify also arbitrarily the circuiting direction (see Fig. 1). We move along the direction of circuiting of one of the contours, If for some intersection the point at which we are located at a given instant is an underpassing point, then this intersection is called an underpass, and in the other case it is called an overpass. We renumber all the underpasses contained on the contour S, underpass No. 1 being taken to be the first underpass encountered on moving from the point zero along the contour. We denote the number of underpasses on the contour S by M. Part of the contour, contained between the $(k-1)$-st and k-th underpasses is called the k-th generatrix and is designated x_k (see

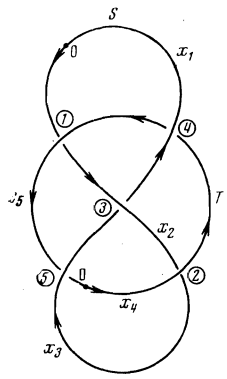

FIG. 1. Example of linkage, shown in regular position, with all the underpasses and generatrices (M = 3, N = 5).

Fig. 1). The generatrix lying between the last and the first underpasses of the contour S is designated x_1. We assign to the first underpass of the contour T the number M + 1 and number in succession all the remaining underpasses of this contour. Its generatrices are designated analogously (see Fig. 1). If N is the total number of underpasses in both contours (i.e., the total number of the intersection points), then the generatrix lying between underpasses M and M + 1 is designated x_{M+1}.

Underpasses can be of two types (I and II), depending on the direction of the overpassing generatrix (see Fig. 2). Thus, just as in the case of a knot, the regular position of the linkages characterized by a sequence of underpasses, for each of which we indicate its type (I or II) and the number of the overpassing generatrix, the overpassing generatrix x_i belonging to the contour S if $i \leq M$ and to the contour T if $i > M$. This description of the regular position can be represented in the form of a square matrix of rank N, called the Alexander matrix for the linkage. The k-th row of this matrix corresponds to the k-th underpass and consists of N elements a_{kj}. These elements are determined in accordance with the following rules[1] (it is assumed that all the elements of the row for which their values are not indicated are equal to zero; i is the number of the overpassing generatrix for the k-th underpass):

1) $k < M$; a) for $i = k$ or $i = k + 1$
   $a_{kk} = -1$, $a_{kk+1} = 1$ independent of the type of underpass;
   b) for $i \neq k$, $i \neq k + 1$, $i \leq M$
   $a_{kk} = 1$, $a_{kk+1} = -s$, $a_{ki} = s - 1$ for underpass of type I,
   $a_{kk} = -t$, $a_{kk+1} = 1$, $a_{ki} = s - 1$ for underpass of type II;
   c) for $i > M$
   $a_{kk} = 1$, $a_{kk+1} = -t$, $a_{ki} = s - 1$ for underpass of type I,
   $a_{kk} = -t$, $a_{kk+1} = 1$, $a_{ki} = s - 1$ for underpass of type II.
2) $k = M$; a) for $i = k$ or $i = 1$
   $a_{kk} = -1$, $a_{k1} = 1$ independent of the type of underpass;
   b) for $i \neq k$, $i \neq 1$, $i \leq M$
   $a_{kk} = 1$, $a_{k1} = -s$, $a_{ki} = s - 1$ for underpass of type I,
   $a_{kk} = -s$, $a_{k1} = 1$, $a_{ki} = s - 1$ for underpass of type II;
   c) for $i > M$, $M > 1$
   $a_{kk} = 1$, $a_{k1} = -t$, $a_{ki} = s - 1$ for underpass of type I,
   $a_{kk} = -t$, $a_{k1} = 1$, $a_{ki} = s - 1$ for underpass of type II;
   d) for $i > M$, $M = 1$
   $a_{kk} = 1 - t$, $a_{ki} = s - 1$ independent of the type of underpass.
3) $k > M$[2]; a) for $i = k$ or $i = k + 1$
   $a_{kk} = -1$, $a_{kk+1} = 1$ independent of the type of underpass;
   b) for $i \neq k$, $i \neq k + 1$, $i > M$
   $a_{kk} = 1$, $a_{kk+1} = -t$, $a_{ki} = t - 1$ for underpass of type I,
   $a_{kk} = -t$, $a_{k,k+1} = 1$, $a_{ki} = t - 1$ for underpass of type II;
   c) for $i \leq M$, $N > M + 1$
   $a_{kk} = 1$, $a_{kk+1} = -s$, $a_{ki} = t - 1$ for underpass of type I,
   $a_{kk} = -s$, $a_{kk+1} = 1$, $a_{ki} = t - 1$ for underpass of type II,
   d) for $i \leq M$, $N = M + 1$
   $a_{kk} = 1 - s$, $a_{ki} = t - 1$ independent of the type of underpass.

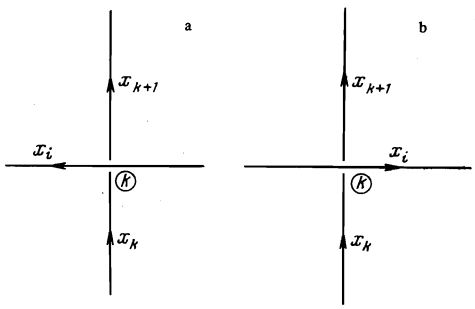

FIG. 2. Two types of underpass: a) type I, b) type II.

We calculate next any minor A_{kj} of order N − 1 of the Alexander matrix and divide it by $s - 1$ if $j \leq M$ and by $t - 1$ if $j > M$. The resultant expression is multiplied by $\pm t^{-m}s^{-n}$ (m and n are integers), so that the resultant polynomial has no negative powers, and the positive powers are minimal, and the term with the largest total exponent must be positive. The polynomial $\Delta(s, t)$ defined in this manner is called the Alexander polynomial for the linkage of two contours. It is an invariant–it is rigorously proved in linkage theory that the Alexander polynomials $\Delta(s, t)$ coincide for equivalent linkages.

Linkages can be classified in analogy with the classification of the knots, in accordance with the minimum number of intersections on their projections. Linkages with equal number of intersections are arranged in a certain definite sequence, and the corresponding serial number is used as an index for the number of intersections. Unfortunately, we were unable to find in the literature a table of even the simplest linkages[3]. We have compiled a table, which to be sure is far from complete, of the simplest linkages, and its initial part is shown in Fig. 3. The Alexander polynomials corresponding to these linkages are given in Table I. Fig. 3 shows only simple linkages, but they can be used to form composite linkages, which are obtained if on one of the chains one ties on a knot in such a way that it is not intersected by another chain. The Alexander polynomial of a composite linkage is equal to the product of the polynomial of the simple linkage and the Alexander polynomial for the additional introduced knot. In spite of the fact that the Alexander polynomial is not a complete invariant (there exist nonequivalent linkages having identical polynomials 5_1 and 7_2 in Table I), it offers definite advantages over the Gauss invariant, which takes the form (see, e.g., [6,10])

$$G = \frac{1}{4\pi} \oint\limits_{S} \oint\limits_{T} \frac{[dr_s\, dr_t] r_{st}}{r_{st}^3},$$

where $r_{st} = r_s - r_t$; r_s and r_t are the radius vectors of the points lying on the contours S and T. There exists a simple connection between the Alexander polynomial and the Gauss integral (see[11]):

$$|G| = \Delta(1,1).$$

We see therefore that the Gauss invariant contains much less information than the Alexander polynomial $\Delta(s, t)$. Thus, for example, we have T = 0 for the linkages 5_1, 7_2, 7_3, and 7_5 (see Table I), i.e., the Gauss invariant does not distinguish all these cases from the case of unlinked contours. At the same time, the Alexander

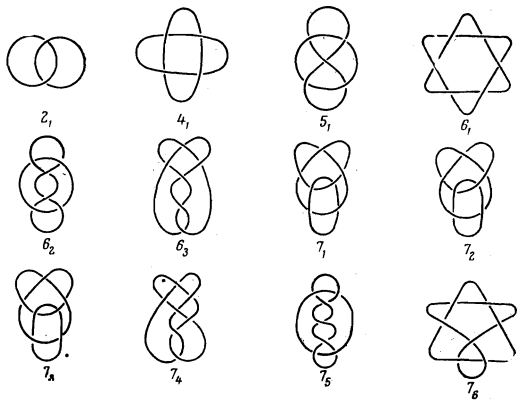

FIG. 3. Linkages with less than eight intersections. The corresponding Alexander polynomials are listed in Table I.

TABLE I. Alexander polynomials $\Delta(s, t)$ for the linkages shown in Fig. 3.

Type of linkage	$\Delta(s, t)$	Type of linkage	$\Delta(s, t)$
2_1	1	7_1	s^3+t
4_1	$s+t$	7_2	$(s-1)(t-1)$
5_1	$(s-1)(t-1)$	7_3	$s^3t-s^3-s^2t+st+s^2-(s+t)+1$
6_1	s^2+t^2+st	7_4	$st(s+t)-(s+t)^2-st+s+t$
6_2	$2st-(s+t)+2$	7_5	$2(s-1)(t-1)$
6_3	$st(s+t)-st+s+t$	7_6	$s^2t^2-st(s+t)+st-(s+t)+1$

Note. For non-linked contours $\Delta(s, t) \equiv 0$. The form of the Alexander polynomial is determined accurate to the substitution $s \to 1/s$ of $t \to 1/t$, so that for example, the polynomials $s^2 + t^2 + st$ and $s^2t^2 + st + 1$ are regarded as equivalent.

polynomial turns out to be different from zero for all the linkages which we have constructed (about 20). The Alexander polynomial $\Delta(s, t)$ vanishes identically for unlinked contours and is equal identically to unity for the simplest linkage 2_1. For all the constructed more complicated linkages it turned out that $|\Delta(-1, -1)| \geq 2$. At the same time, in the calculations performed in the present study we are interested in whether a considered pair of closed chains belongs to only one of three classes: the absence of a linkage, the simplest linkage 2_1, and all the remaining complex linkages. Within the framework of this problem, we confine ourselves to the calculation of only $\Delta(-1, -1)$.

CALCULATIONS OF TOPOLOGICAL INTERACTION

We consider two polymer molecules. We assume that the interaction between the chains or between segments that are far separated along a chain consists of only in the impossibility of the passing through each other. The chains will be considered here to be equal to infinitesimally thin. If the molecules are not closed, then the states of any one of them does not depend in any way on the state of the other and there is no interaction between the molecules at all. Closing one of the chains does not change the situation. If both chains are closed, then an entropy-type interaction will appear between them. In fact, when closed, the molecules turn out to be in a certain definite topological state (the absence of linkage or linkage of any one type, see Fig. 3), which subsequently should be preserved for any possible chain deformation. This topological limitation leads to a decrease of the conformation entropy of polymer molecules.

The degree of this decrease, and consequently also the free energy of the system, depends on the topological state of the chains and on the distance between them.

Thus, to find the topological interaction between two closed polymer molecules that are in a given topological state it is necessary to calculate the dependence of the number of conformations of the chains on the distance between them. To this end it is necessary, first of all, to have an invariant that distinguishes between a topological state of a system consisting of two closed chains. We have used in this connection the algebraic invariant (the Alexander polynomial) described in the preceding section.

The invariant $\Delta(s, t)$ employed for the linkages is a polynomial that depends on two variables, s and t. Equivalent linkages always give equivalent Alexander polynomials. The examples of polynomials obtained for linkages of different types shown in Fig. 3 are given in Table I. In the absence of a linkage we have $\Delta(s, t) \equiv 0$.

The number of conformations of chains that are in a given topological state is the product of the total number of conformations of the isolated chains and the probability of formation of a given topological state for random closing of the chains.

To find the probability of formation of a given topological state, we used the Monte Carlo method. To this end, we generated independently and randomly two closed polymer chains of definite length L (expressed in terms of the number of segments) on a body-centered lattice. The construction of each of the chains was by the method described in the earlier paper[10]. In the present study we confined ourselves to consideration of only a model of an infinitesimally thin chain. In this model, the polymer molecule is regarded as a sequence of infinitesimally thin segments (of a straight line), each of which can assume, relative to the preceding segment, seven independent directions on the body-centered lattice (a backward step is forbidden).

For each of the two chains, we determined the mass-center coordinates[4)], and the chains were then shifted in such a way that the distance R between their mass centers assumed a specified value. This operation was effected by a simple parallel transfer without any deformations of the chain, and one chain was allowed to pass unobstructed through another. After completing this operation of chain displacement, the topological state of the produced system was assessed with the aid of the invariant described in the preceding section. Within the framework of this paper we confined ourselves to breaking up all the possible topological states into three classes–absence of linkage, simplest linkage 2_1 (see Fig. 3), and more complicated linkages.

The calculations were performed with a BÉSM-6 computer. The lengths L for which the calculations were performed were L = 20–80 segments. Figure 4 shows the results of computer calculations of the dependence of the probabilities of the absence of linkage (P_0) on the distance the mass centers of the chains. Generally speaking, the sought probabilities should depend on the topological state of each of the two considered chains (i.e., on the type of the knot–trivial knot, trefoil knot, etc.). We could verity whether a dependence of this kind is significant by using the knot-classification algorithm described in our preceding paper[10]. The verification has shown that in the chain-length region investigated in the present paper the quantity P_0 is independent of the topological state of each of the chains, within the limits of the statistical calculation error.

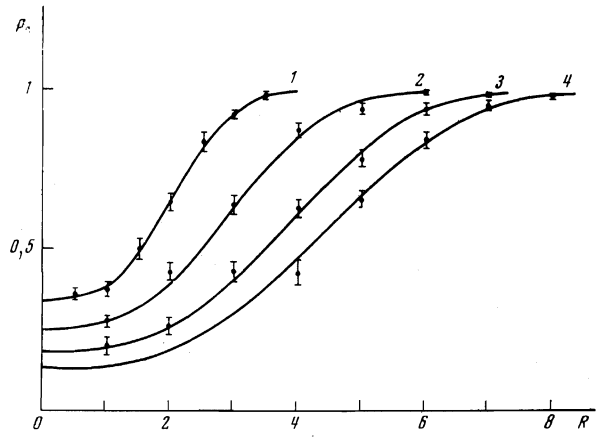

FIG. 4. Dependence of the probability of absence of linkage in the case of random closing of two chains, on the distance R between their mass centers. The different curves correspond to different chain lengths L: 20 (curve 1), 40 (curve 2), 60 (curve 3), and 80 (curve 4). The values of R and L are given in terms of the number of segments. Points – results of computer calculations. The curves are plots of formula (1).

An analysis of the results of the calculations of the probability P_0 has led to an unexpected result. It turned out that for all L for which the calculations were performed the value of P_0 exhibited an identical dependence on R, of surprisingly simple form:

$$P_0=1-A_0\exp(-\alpha_0 R^3). \qquad (1)$$

The extent to which (1) agrees well with the results of the calculations can be assessed from Fig. 4, where the curves are plots of formula (1) and the points correspond to the results of computer calculations. Table II lists the values of the parameters A_0 and α_0 for different L.

We note that A_0 depends very little on L and tends to unity with increasing L. To the contrary, α_0 depends significantly on L ($\alpha_0 \propto L^{-1.7}$ in the considered length interval).

In calculations of the probability of the formation of the linkage 2_1 we can no longer disregard the topological state of each of the chains in the system. It is natural to confine oneself to unknotted chains (i.e., chains forming a trivial knot). The results of the corresponding calculations are shown in Fig. 5. These results can no longer be described with the aid of a sufficiently simple and universal interpolation formula, For small lengths (L = 20 and L = 40), however, the following formula holds approximately true:

$$P_2\approx A_2\exp(-\alpha_2 R^3). \qquad (2)$$

The curves plotted in accordance with formula (2) are given in Fig. 5.

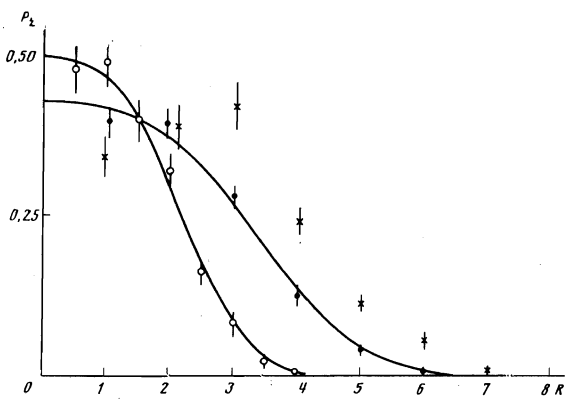

FIG. 5. Probability of formation of the simplest linkage 2_1 following random closing of two chains, vs distance R between their centers. Points – results of computer calculations: ○ -- L = 20, ● -- L = 40, X -- L = 60. The curves are plots of formula (2) for the following values of the parameters: A_2 = 0.5 and α_2 = 0.64 at L = 20; A_2 = 0.43 and α_2 = 0.018 at L = 40.

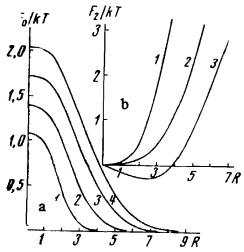

FIG. 6. "Potentials" of topological interaction for non-linked polymer molecules (a) and for molecules forming the simplest linkage 2_1 (b). The curves are plotted in accordance with formula (3) and the results shown in Figs. 4 and 5 for different values of chain links SL: 20 (curve 1), 40 (curve 2), 60 (curve 3), and 80 (curve 4).

TABLE II. Parameters of the interaction "potential" between non-linked polymer molecules (α_0 and A_0), and values of the second virial coefficient B for different lengths L.

L	α_0	A_0	B	$^1/_{12}\pi L^{3/2}$	L	α_0	A_0	B	$^1/_{12}\pi L^{3/2}$
20	$8.2\cdot10^{-2}$	0.66	17	23	60	$1.2\cdot10^{-2}$	0.82	143	121
40	$2.6\cdot10^{-2}$	0.75	60	66	80	$0.8\cdot10^{-2}$	0.87	228	185

The free energy F_i of the system in a given topological state i is connected with the obtained probabilities by the relation

$$F_i=-kT\ln P_i. \qquad (3)$$

The dependence of the free energy of the system on the distance between the chains, which can be regarded as the "potential" of the chain interaction, is shown in Fig. 6 for two simplest topological states.

The presence of a repelling "potential" for the interaction between the non-linked closed molecules can become manifest in the presence of the second virial coefficient for a gas or a solution of closed polymer chains. This quantity can be calculated from the standard formula (see, for example,[14])

$$B=\frac{1}{2}\int\left[1-\exp\left(-\frac{F_0}{kT}\right)\right]dV. \qquad (4)$$

Using formulas (1) and (3), we obtain from (4)

$$B=^2/_3\pi A_0/\alpha_0. \qquad (5)$$

The values of B calculated for different L are shown in Table II. For comparison, the last column of the table shows the values of the second virial coefficient for a hypothetical system of hard spheres of diameter $(\overline{h^2})^{1/2}$, where h is the distance between two farthest points, along the chain, of a closed polymer molecule consisting of L fully independent segments[5)].

DISCUSSION OF RESULTS

The calculations show that allowance for the topological limitations in the analysis of the statistical properties of two closed polymer chains gives rise to an entropy-type interaction between them. This interaction turned out to be quite appreciable and leads to the fact that in practice two non-linked polymer coils can not penetrate into each other. With increasing distance R between the non-linked polymer coils, the "potential" of the interaction between them decreases like (see formulas (1) and (3))

$$F_0=-kT\ln[1-A_0\exp(-\alpha_0 R^3)] \qquad (6)$$

The dependence of the parameters A_0 and α_0 on the

length of the polymer chains is given in Table II. The form of this "potential" was shown in Fig. 6a. The interaction "potential" between polymer chains that are in the simplest linked state 2_1 has an entirely different form (see Fig. 6b) and in the case of small L it follows approximately the simple law[6)]

$$F_2 \approx kT\alpha_2 R^3. \quad (7)$$

These results illustrate the essential dependence of the interaction "potential" between chains on the character of their topological states.

The results are of interest in connection with a number of questions, some of which we shall consider briefly.

1. Effects of topological interaction can greatly influence the behavior of DNA molecules in the helix-coil transition interval, for owing to the two-filament character of the DNA molecule its melted section, which is surrounded by non-melted sections, constitutes a closed polymer chain. This circumstance was pointed out in[15], where it was shown that to interpret the experimental data on the characteristic viscosity of partially melted DNA it is necessary to assume that the coil-like sections produced during the course of melting cannot penetrate into one another. The calculations of the present paper confirm the possibility of such an interpretation.

2. There exist unique chemical compounds that constitute linkages of two or more closed polymer molecules and called catenanes (see[16]). The main method of obtaining catenanes is random closing of linear polymer chains. The question of the probability of producing catenanes by this method has been discussed in a number of papers[16-20].

The concentration C_C of the catenanes produced following random closing of chains with concentration C is equal to

$$C_K = C^2 B, \quad (8)$$

where B is given by (4). Formula (8) is valid because we assume the molecules of the polymer not to interact with one another up to the instant when both chains are closed, under the condition $CB \ll 1$. Thus, the second virial coefficient for closed non-linked molecules is at the same time the equilibrium constant of the catenane-production reaction.

Quantitative estimates of the catenane-production probability were first made by Frisch and Wasserman[18]. In their calculations, they assumed that the probability of catenane production is proportional to the volume occupied by the polymer coil. Our calculations show that this assumption is indeed approximately valid. Moreover, it is quite curious that Frisch and Wasserman also chose successfully the proportionality coefficients in such a way that the formula gives for B numerical values that agree quite satisfactorily with the results of our calculations.

In quantitative estimates of the probability of catenane production it must be recognized that our results pertain to a model with an absolutely thin chain. The closest to this idealized model is the two-filament DNA molecule, for which the ratio of the segment length to its thickness is very large (≈ 50). The DNA molecules form various linkages both under artificial conditions and, in certain pathological cases, under natural conditions (see[19, 20]).

Wang and Schwartz[19] made a rough experimental estimate of the probability of formation of a linkage between two DNA molecules under random-closing conditions. However, there are at present unfortunately no quantitative experimental data on this process.

3. As is well known, the elasticity of polymer systems is of entropy origin. The validity of Hooke's law for such systems is to a certain degree accidental–it follows from the Gaussian character of the distribution of the distances between the ends of the polymer chain (see, e.g.,[21]). Our results show that polymer systems constructed on the basis of catenanes may not obey Hooke's law. In fact, it follows from (7) that the force for catenanes may depend on the strain in a nonlinear manner.

The authors are grateful to Yu. S. Lazurkin for interest in the work and for useful discussions.

1)It is assumed that on each of the contours there is at least one underpass.
2)The relations presented in this item are valid if the substitution $N + 1 \rightarrow M + 1$ is made.
3)Such a table does exist for knots [13] (it contains all the knots with less than 10 intersections).
4)It was assumed here that the masses are concentrated on the ends of the segments.
5)It should be borne in mind that since a step backward is forbidden in our model, the length of the chain L does not correspond, strictly speaking, to the number of statistical segments in it, which is in fact equal to $\sqrt{3}L/2$.
6)We have discarded in (7) the free-energy term that does not depend on R.

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