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Linking probabilities of off-lattice self-avoiding polygons and the effects of excluded volume

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Abstract
We evaluate numerically the probability of linking, i.e. the probability of a given pair of self-avoiding polygons (SAPs) being entangled and forming a nontrivial link type L. In the simulation we generate pairs of SAPs of N spherical segments of radius r_d such that they have no overlaps among the segments and each of the SAPs has the trivial knot type. We evaluate the probability of a self-avoiding pair of SAPs forming a given link type L for various link types with fixed distance R between the centers of mass of the two SAPs. We define normalized distance r by $r = R/R_{g,0_1}$ where $R_{g,0_1}$ denotes the square root of the mean square radius of gyration of SAP of the trivial knot 0_1. We introduce formulae expressing the linking probability as a function of normalized distance r, which gives good fitting curves with respect to χ^2 values. We also investigate the dependence of linking probabilities on the excluded-volume parameter r_d and the number of segments, N. Quite interestingly, the graph of linking probability versus normalized distance r shows no N-dependence at a particular value of the excluded volume parameter, $r_d = 0.2$.

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1. Introduction

Topological effects among ring polymers have attracted much attention recently in various research fields such as DNA, proteins and synthetic polymers [1–4]. The topology of a ring polymer is expressed by a knot type and is kept unchanged under thermal fluctuations once the ring polymer is formed. Quite interestingly, topological constraints of ring polymers may lead to nontrivial statistical mechanical and dynamical properties of ring polymers (see for instance [5] and references therein).

Two simple closed curves can be mutually entangled. The topology of a pair of closed curves is described by a link type. Some links are depicted in figure 1. In this paper, we

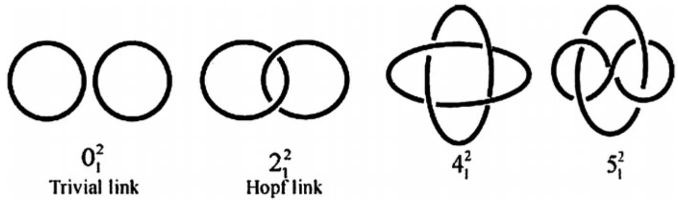


Figure 1. Trivial link 0_1^2 and some nontrivial links. Symbol 2_1^2 denotes the Hopf link.

discuss topological interaction between such two ring polymers in solution that have the trivial knot type. In particular, we numerically evaluate the probability of two ring polymers forming a link type which consists of two trivial knots when they are synthesized randomly in good solvent [6–8]. We model a ring polymer as a self-avoiding polygon (SAP) consisting of N hard spherical beads of radius r_d with unit bond length. We construct a large number of self-avoiding pairs of SAPs of N beads where the centers of mass of the two SAPs are separated by a fixed distance R. Here we assume that in each self-avoiding pair of SAPs there is no overlap among the $2N$ beads. We also assume that each SAP has the trivial knot type. We define the linking probability, $P_{\rm link}(R, N, r_d)$, by the probability for such a self-avoiding pair of SAPs of the trivial knot having the topology of a nontrivial link. If the link is nontrivial, then the two polygons are mutually entangled so that they cannot be separated by continuous mapping of the three-dimensional space. More precisely, for such a link type L that consists of two trivial knots we define the linking probability of link L by the probability that a given self-avoiding pair of SAPs has the link type L where the centers of mass is separated by R. We denote it by $P_L(R, N, r_d)$. Here we recall that each SAP has the trivial knot type. In terms of this notation, $P_{\rm link}$ is given by $P_{\rm link} = 1 - P_{0_1^2}$, where 0_1^2 denotes the trivial link of two components (see figure 1).

The linking probability is relevant to the osmotic pressure of a dilute solution of ring polymers. By measuring the osmotic pressure [9] it was shown that the second virial coefficient of a ring-polymer solution does not vanish at the theta temperature of the solution of the corresponding linear polymers. We call it the anomalous second virial coefficient at the theta temperature of the linear polymer, θ_l, and denote it by $A_2(\theta_l)$. It is given by

$$A_2(\theta_l) = N_A/(2M^2) \int_0^\infty P_{\rm link}(R, N, 0) 4\pi R^2 \, {\rm d}R, \tag{1}$$

where M denotes the molecular weight of a ring polymer and N_A is Avogadro's number [6, 10, 11]. The $A_2(\theta_l)$ was explained theoretically first by evaluating the linking probability through the topological moment [10, 11], which is the expectation value of the linking number between two random polygons [12]. However, the theoretical results were not completely consistent with the experimental data. In fact, the linking number is only a homological invariant of two spatial curves so that the topological moment only approximately expresses the topological entropic constraints for pairs of ring polymers in solution.

In this paper, we numerically evaluate linking probabilities of two SAPs of the trivial knot type, $P_{\rm link}(R, N, r_d)$ and $P_L(R, N, r_d)$ for various link types L consisting of two trivial knots, and introduce a formula for expressing them as a function of distance R between the centers of mass of given two polygons. Here we emphasize that the formula gives good fitting curves also from the viewpoint of the χ^2 values. There are several numerical studies on linking probabilities [6–8, 13]. However, no fitting formulae in the previous studies are consistent with the data with respect to the χ^2 values. Furthermore, we also investigate the r_d-dependence and the N-dependence of linking probabilities, and show their nontrivial properties such as in the case when the excluded volume parameter is large.

The linking probability was numerically evaluated first for random polygons on the cubic lattice for $N \leqslant 80$ and a prototype of the fitting formula was introduced [6]. Here an ensemble of random polygons was regarded as a simple model of circular DNA. For circular DNA N is limited such as $N \leqslant 50$ in experiments. However, for synthetic ring polymers in solution N can be as large as $N = 10^3$. By combining different link invariants, the linking probability of random polygons has been evaluated for $N \leqslant 500$, and the N-dependence of $A_2(\theta_l)$ is consistent with the simulation result [8]. A good fitting formula of the linking probability for link type $L = 2_1$ was also obtained [8]. Here we recall that random polygons have no excluded volume. The linking probability was evaluated for SAP consisting of cylindrical segments of unit length with radius $r_d = 0.015$ for $N = 20$ [7], and for SAP consisting of hard beads of radius r_d for upto $N = 257$ and $r_d = 0.3$ [13]. Different fitting formulae were introduced in [7] and [13]. However, the fitting curves were not good with respect to the χ^2 values.

Different definitions of the linking probability have been studied numerically or theoretically. The linking probability of lattice polygons confined in a finite volume was studied rigorously and numerically through lattice simulation [14]. The linking number of two spatial curves has been generalized [15]. The probability of linking in higher dimensions has also been studied rigorously [16]. Furthermore, some rigorous results on random knotting of theta curves are obtained recently [17], which can be considered as a generalization of random linking of two loops.

Linking probability should also be important in DNA. Linking of DNA chains is particularly relevant in chromosome biology [18]. The end products of replication in *Escherichia coli* are two linked circles. Resolution of this topological problem is essential to ensure cell division. In association with the linking of DNA chains, linking probability of uniform random polygons in confined space has been studied rigorously and numerically [19].

We define normalized distance r by $r = R/R_{g,0_1}$ where $R_{g,0_1}$ denotes the square root of the mean square radius of gyration of SAP of the trivial knot 0_1. Hereafter, we use the normalized distance r instead of R, i.e. we denote $P_L(R_{g,0_1}r, N, r_d)$ simply by $P_L(r, N, r_d)$, and we call the normalized distance r simply distance r.

The content of this paper consists of the following. In section 2 we introduce simulation methods and fundamental algorithm of the present research, i.e. that of generating self-avoiding pairs of SAPs forming a given link type. In section 3, we numerically evaluate the mean square radius of gyration of self-avoiding pairs of SAPs. The simulation result supports at least partially the validity of the algorithm formulated in section 2. In section 4, we introduce a fitting formula for the linking probability of link type L as a function of distance r between the centers of mass of a given self-avoiding pair of SAPs. Applying it to the simulation data we obtain good fitting curves with respect to χ^2 values. We discuss an intuitive derivation of the formula. In section 5, we discuss the r_d-dependence and the N-dependence of the linking probability. In section 6 we give conclusions.

2. Simulation methods

2.1. Model of SAP

Let us introduce the rod-bead model of SAP [20]. A conformation of the SAP is given by a sequence of N line segments of unit length. A solid ball of radius r_d is placed at each vertex of the polygon and the balls are not allowed to intersect each other for a valid SAP.

In the simulation of random linking we consider only such SAPs that have the trivial knot type.

2.2. Algorithm for generating SAP

To construct random conformations of off-lattice SAP, we introduce the crank-shaft moves and the bond-interchange moves. Here we note that the former moves are similar to some parts of the pivot moves for generating self-avoiding polygons on a lattice [21].

The crank-shaft algorithm consists of the following procedures. First we take a conformation of SAP in three dimensions. Second, we choose randomly two nodes of the SAP, and we rotate a subchain between the nodes around the axis passing through them. Here the rotation angle θ ($0 \leqslant \theta < 2\pi$) is chosen randomly with uniform probability. If there is an overlap among segments in the rotated conformation, then we throw it away and return to the initial conformation. If the rotated conformation has no overlaps among segments, we accept it as a valid SAP. We repeat the procedure until the chain reaches a state which is effectively independent of the initial one.

In addition to the crank-shaft moves, we also apply the bond-interchange moves, by which possible local correlations may decrease rapidly between the initial and the deformed conformations of SAP [22, 23]. In every bond-interchange move, we choose two bonds randomly among all bonds of a given polygon and then interchange them, and search possible overlaps among the segments of the new conformation. If there is no overlap, we accept the interchanged conformation as a valid SAP. If they have an overlap, we throw it away and return to the initial conformation. Through the moves, the alignment of bonds is rearranged while the directions of the bonds are preserved.

In the simulation we perform the crank-shaft and the bond-interchange moves $2N$ and N times, respectively, and we make a new SAP. In fact, according to previous studies, it is appropriate to perform the crank-shaft moves at least N times for obtaining such a new conformation that is effectively independent of the initial one [24]. For lattice polygons, it was shown that the correlation with the initial conformation decreases with respect to the number of applied moves [24].

After constructing a large number of SAPs we calculate the second order Vassiliev invariant and the determinant of knots for the SAPs. We then select such SAPs that have the same set of values of the two knot invariants with the trivial knot. We thus practically select the SAPs of the trivial knot among the large number of generated SAPs.

2.3. Construction of links of self-avoiding polygons

We define a link by a set of more than or equal to two components of polygons or closed curves in three dimensions. Two sets of polygons or curves have the same link type if and only if one of the sets is derived from the other set by a continuous map of the three-dimensional space. If the two sets of polygons or curves have the same link type, we say that they are topologically equivalent. A link is called *trivial* if all the components can be separated by a continuous map of the three-dimensional space where no segments of the polygons or the curves cut or cross each other. If a link is not trivial, it is called a nontrivial link. In this paper, we only consider those links whose components are the trivial knot.

Let us now consider an ensemble of pairs of SAPs in which each SAP consists of N hard spherical beads with radius r_d such that each of the pairs makes a given link type L and has no overlaps among the segments. Here we assume that in each pair the knot types of the SAPs are trivial, i.e. each component of the link L has the trivial knot type. We call such a pair of SAPs a *self-avoiding pair of SAPs of link type L* or a *self-avoiding link L of SAPs*, for short.

Table 1. Values of the two link invariants, the linking number and the Alexander polynomial evaluated at $t = -1$ for some simple two-component links.

Link type	0_1^2	2_1^2	4_1^2	5_1^2	6_1^2	6_2^2	6_3^2
Linking number	0	1	2	0	3	2	3
Alexander polynomial	0	1	2	4	3	6	5

Furthermore, we construct such an ensemble of self-avoiding links of SAPs where the centers of mass of each pair of SAPs are separated by the same distance r. Here we recall that symbol r denotes the normalized distance with respect to the square root of the mean square radius of gyration of the SAP of the trivial knot 0_1, $R_{g,0_1}$, i.e. we have $r = R/R_{g,0_1}$.

Let us formulate the procedure for generating an ensemble of self-avoiding links of SAPs where each SAP has N beads of radius r_d. Here we note that the derived link has $2N$ segments. Suppose that we have an ensemble of SAPs of N hard beads of radius r_d. In our simulation we construct 100 000 independent SAPs of the trivial knot type. First, we choose a pair of SAPs among the ensemble randomly. Second, we put them in such a way that there is a distance r between the centers of mass of the SAP. Then, for all pairs of beads, we check whether they overlap or not. If all the pairs have no overlap, the pair of polygons makes a valid self-avoiding link of SAP. Then, we save its conformation data on computer memory, and return to the beginning. If there is an overlap then we throw away the pair of SAPs, and return to the beginning and pick up another pair of SAPs.

We repeat this process a large number of times and we construct an ensemble of self-avoiding links of SAP. In our simulation, we obtain 100 000 links by the method, eventually.

2.4. *Definition of linking probabilities*

Let us systematically define the linking probabilities. We define the *linking probability* of two SAPs of the trivial knot, $P_{\text{link}}(r)$, by the probability that a given self-avoiding pair of SAPs of the trivial knot where the centers of mass of the SAPs are separated by a distance r is topologically equivalent to a nontrivial link.

If a given self-avoiding pair of SAPs is topologically equivalent to the trivial link, we call it a *trivial link* of SAPs, otherwise we call it a *nontrivial link* of SAP. In a nontrivial link of SAP the two polygons are entangled with each other (see figure 1). Here we have assumed that each of the two SAPs has the trivial knot type.

Let L denote such a link that consists of two trivial knots. We define the linking probability of link type L, $P_L(r)$, by the probability that a given self-avoiding pair of SAPs of the trivial knot with distance r between the centers of mass is topologically equivalent to link type L.

The linking probabilities are functions of the number of segments N and the radius r_d of hard spherical beads. Therefore, we also denote $P_L(r)$ and $P_{\text{link}}(r)$ more specifically by $P_L(r, N, r_d)$ and $P_{\text{link}}(r, N, r_d)$, respectively.

Through simulation we numerically evaluate linking probabilities of SAP of the trivial knot. We assume that all configurations of self-avoiding links of SAPs with the trivial knot have statistically the same probability of appearance. We thus calculate the linking probabilities by

$$P_i(r, N, r_d) = \frac{M_i}{M}, \qquad i = L \quad \text{or} \quad \text{link}, \tag{2}$$

where L is a link consisting of two trivial knots, and M_L and M_{link} are the number of self-avoiding links with link type L and that of nontrivial links, respectively. Here M is the total number of self-avoiding links of SAP of the trivial knot generated in the simulation. We recall that $M = 100\,000$ in our simulation.

Calculating link invariants we can practically determine the link type of a given configuration of the self-avoiding link. We use two link invariants called the *linking number* and the *Alexander polynomial* evaluated at $t = -1$. For some simple links we can distinguish them by the values of the two link invariants (see table 1). Thus, if the fraction of rather complicated links is small, we can evaluate the linking probabilities quite accurately.

Hereafter in this paper, we abbreviate the superscript 2 in the symbols of links. For instance, we denote the Hopf link 2_1^2 simply by 2_1.

3. Mean square radius of gyration of self-avoiding links of SAP

In order to check the validity of the algorithm for generating self-avoiding links of SAP, we now numerically evaluate the mean square radius of gyration for self-avoiding pairs of SAPs.

3.1. Mean square radius of gyration of SAPs

Let us first consider an ensemble of SAPs consisting of N segments of hard spherical beads with radius r_d. Suppose that we can select such SAPs from the ensemble that have the trivial knot type 0_1. Then, we take the average of some quantity A over all the conformations of SAPs of the trivial knot 0_1. We denote it by the symbol $\langle A\rangle_{0_1}$. Let $\boldsymbol{r}_j$ for $j = 1, 2, \ldots, N$ be the position vectors of the vertices of a given SAP. We define $R^2_{g,N}$ by

$$R^2_{g,N} = \frac{1}{2N^2}\sum_{j=1}^{N}\sum_{k=1}^{N}(\boldsymbol{r}_j - \boldsymbol{r}_k)^2. \tag{3}$$

Then, the mean square radius of gyration of SAP with the trivial knot type 0_1 is given by the following:

$$\begin{aligned} R^2_{g,0_1} &= \left\langle R^2_{g,N}\right\rangle_{0_1} \\ &= \frac{1}{2N^2}\sum_{j=1}^{N}\sum_{k=1}^{N}\langle(\boldsymbol{r}_j - \boldsymbol{r}_k)^2\rangle_{0_1}. \end{aligned} \tag{4}$$

The square root of the mean square radius of gyration, $R_{g,0_1}$, is given by $R_{g,0_1} = \sqrt{\left\langle R^2_{g,N}\right\rangle_{0_1}}$. Here we note that equation (4) is also expressed as follows:

$$R^2_{g,0_1} = \frac{1}{N}\sum_{j=1}^{N}\langle(\boldsymbol{r}_j - \boldsymbol{r}_{G,0_1})^2\rangle_{0_1},$$

where $\boldsymbol{r}_{G,0_1} = \sum_{k=1}^{N}\boldsymbol{r}_k/N$ denotes the center of mass of the SAP.

The estimates of the mean square radius of gyration of SAP with the trivial knot type are plotted against the number of segments, N, in figure 2. The fitting curves are given by the formula

$$R^2_{g,0_1} = CN^{2\nu}. \tag{5}$$

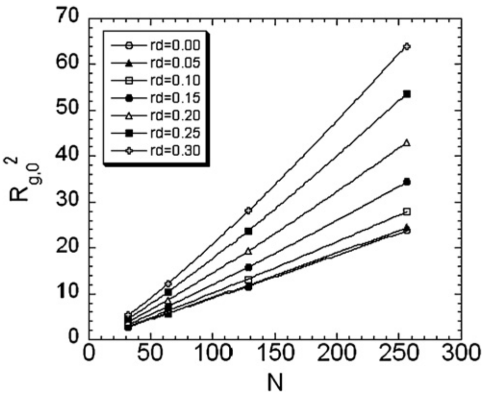


Figure 2. $R^2_{g,0_1}$ versus *N*. We recall that $R^2_{g,0_1}$ denotes the mean square radius of gyration for equilateral SAPs of hard spherical beads with radius r_d having the trivial knot type. The parameters $(C, 2\nu)$ of the fitting curves of formula (5) which are given by (0.073, 1.04) for $r_d = 0$, (0.072, 1.05) for $r_d = 0.05$, (0.068, 1.09) for $r_d = 0.10$, (0.067, 1.13) for $r_d = 0.15$, (0.071, 1.16) for $r_d = 0.20$, (0.077, 1.18) for $r_d = 0.25$ and (0.086, 1.19) for $r_d = 0.30$.

3.2. The estimates of the mean square radius of gyration for self-avoiding links of SAP

Let us now consider a set of self-avoiding pairs of SAPs of *N* segments. Here we do not specify their link types as far as each SAP has the trivial knot 0_1. Suppose that we have a self-avoiding pair of SAPs of *N* segments. We denote by $\boldsymbol{r}_j$ the position vector of the jth vertex of the first polygon for $j = 1, 2, \ldots, N$, and that of the second polygon for $j = N + 1, N + 2, \ldots, 2N$, respectively. Then, the center of mass of the self-avoiding pair, $\boldsymbol{r}_{G,2N}$, is given by

$$\boldsymbol{r}_{G,2N} = \frac{1}{2N}\sum_{j=1}^{2N}\boldsymbol{r}_j. \tag{6}$$

We denote by $R^2_{g,all(0_1,0_1)}$ the mean square radius of gyration for a self-avoiding pair of SAPs where the link type is arbitrary except that each of the SAP should have the trivial knot type 0_1. We define it by the following:

$$\begin{aligned} R^2_{g,all(0_1,0_1)} &= \left\langle R^2_{g,2N}\right\rangle^{0_1,0_1}_{all} \\ &= \frac{1}{2N}\left\langle \sum_{j=1}^{2N}(\boldsymbol{r}_j - \boldsymbol{r}_{G,L})^2\right\rangle^{0_1,0_1}_{all}. \end{aligned} \tag{7}$$

Here the symbol $\langle A\rangle^{0_1,0_1}_{all}$ denotes the average of quantity *A* over all the configurations of the self-avoiding link of SAP of such an arbitrary link type in which each SAP has the trivial knot type 0_1.

The estimates of $R^2_{g,all(0_1,0_1)}$ are plotted against the number of segments, *N*, in figure 3. The fitting curves are given by the same formula (5).

Let us now discuss the relation between the mean square radius of gyration of SAPs of the trivial knot, $R^2_{g,0_1}$, and that of self-avoiding links of SAPs with any link type where each SAP has the trivial knot, $R^2_{g,all(0_1,0_1)}$. We first consider the case of no excluded volume, i.e. $r_d = 0$. We denote by $\boldsymbol{r}_{G,1}$ and $\boldsymbol{r}_{G,2}$ the centers of mass of the first and second polygons, respectively. Then, $R^2_{g,all(0_1,0_1)}$ is expressed in terms of the centers of mass of the two polygons as follows:

$$R^2_{g,all(0_1,0_1)} = \left\langle R^2_{g,N}\right\rangle^{0_1,0_1}_{all} + \tfrac{1}{4}\langle(\boldsymbol{r}_{G,1} - \boldsymbol{r}_{G,2})^2\rangle^{0_1,0_1}_{all}. \tag{8}$$

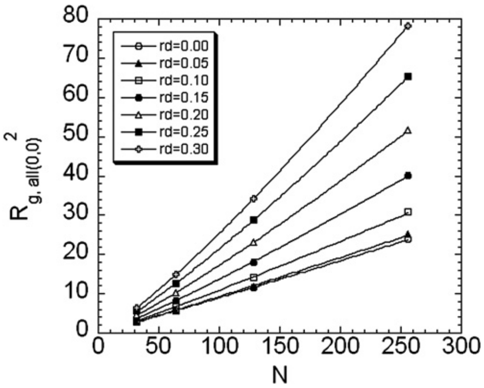


Figure 3. The mean square radius of gyration for self-avoiding pairs of SAPs of hard spherical beads with radius r_d, $R^2_{g,all(0_1,0_1)}$, plotted against *N* for various values of r_d. Here the excluded volume parameter r_d is given by $r_d = 0.0, 0.05, 0.10, 0.15, 0.20, 0.25, 0.30$.

Here the center of mass of the self-avoiding link of SAP is given by

$$\boldsymbol{r}_{G,L} = \tfrac{1}{2}(\boldsymbol{r}_{G,1} + \boldsymbol{r}_{G,2}).$$

If we set $\boldsymbol{r}_{G,1} = \boldsymbol{r}_{G,2} = \boldsymbol{0}$, we obtain

$$R^2_{g,all(0_1,0_1)} = \left\langle R^2_{g,N} \right\rangle^{0_1,0_1}_{all}. \tag{9}$$

Thus, if the centers of mass of random polygons are fixed at the origin, the mean square radius of gyration of pairs of random polygons of the trivial knot, $R^2_{g,all(0_1,0_1)}$, is equal to that of one of the two random polygons of the trivial knot which compose a link of two trivial knots, $\left\langle R^2_{g,N} \right\rangle^{0_1,0_1}_{all}$. The latter corresponds to the mean square radius of gyration of random polygons with the trivial knot, $R^2_{g,0_1}$. Therefore, if the centers of mass of random polygons are fixed at the origin, we have the following equality for random polygons:

$$R^2_{g,all(0_1,0_1)} = R^2_{g,0_1}. \tag{10}$$

In short, if the excluded volume is zero and the centers of mass of random polygons are fixed at the origin, then the self-avoiding links and the SAPs have the same mean square radius of gyration.

If the excluded volume is non-zero, however, the mean square radius of gyration of a self-avoiding link of SAP should be larger than that of the SAP even if the centers of mass of SAPs are fixed at the origin. That is, we should have $R^2_{g,all(0_1,0_1)} > R^2_{g,0_1}$ if the centers of mass of SAPs are fixed at the origin. The average size of self-avoiding links should be larger than that of SAP due to the effective repulsion among segments arising from the excluded volume.

In figure 4, the ratio $R^2_{g,all(0_1,0_1)} \big/ R^2_{g,0_1}$ is plotted against the number of segments of SAP, *N*. We confirm that the ratio is given by 1.0 if the excluded volume is zero ($r_d = 0$), and also that the ratio becomes greater than 1.0 if the excluded volume is non-zero ($r_d > 0$). Furthermore, the *N*-dependence of the ratio is very small. The graph of the ratio versus *N* is almost flat. The numerical result suggests that the algorithm of generating self-avoiding links of SAP should be valid.

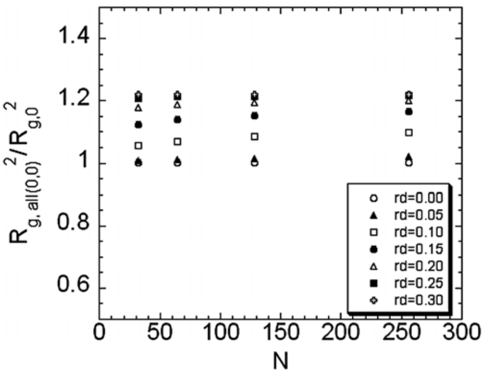


Figure 4. Ratio of the mean square radius of gyration of self-avoiding pairs of SAPs of the trivial knot to that of SAP of the trivial knot, $R^2_{g,all(0_1,0_1)}/R^2_{g,0_1}$, versus the number of segments, N. Here K is given by the trivial knot, and the excluded volume parameter r_d is given by $r_d = 0.0, 0.05, 0.10, 0.15, 0.20, 0.25, 0.30$.

4. Formula of the linking probability

4.1. Good fitting curves with respect to χ^2 values

Let us introduce a fitting formula for the linking probability as a function of distance r between the centers of mass of SAP

$$P_i(r) = \exp(-\alpha r^{\nu_1}) - C\exp(-\beta r^{\nu_2}), \qquad i = L \quad \text{or} \quad \text{link}, \tag{11}$$

where C, α, β, ν_1 and ν_2 are fitting parameters. An intuitive derivation of the fitting formula (11) will be given in subsection 4.2. We note that a similar formula was introduced for the linking probability of the Hopf link where ν_1 and ν_2 are fixed as $\nu_1 = \nu_2 = 3$ [8].

We have applied formula (11) to the data points of $P_{\text{link}}(r)$ and $P_L(r)$ obtained through simulation. The fitting curves are depicted together with the data points in figure 5. Each panel contains five fitting curves for P_{link}, P_{2_1}, P_{4_1}, P_{5_1} and P_{others}, which correspond to the linking probabilities of the following link types: all the nontrivial links, 2_1, 4_1, 5_1, and all the nontrivial links other than 2_1, 4_1 and 5_1, respectively.

The graphs given in figure 5 clearly show that formula (11) gives good fitting curves to the data points for $N = 256$. Similarly, formula (11) also gives good fitting curves to the data points for the cases of $N = 32$, 64 and 128. Furthermore, we have obtained good χ^2 values at least for the cases of $r_d \leqslant 0.20$. Here we remark that there are 31 data points for one fitting curve. The best estimates of the five parameters C, α, β, ν_1 and ν_2 together with the χ^2 values are listed in tables 2 and 3. The χ^2 values are sufficiently small for $r_d \leqslant 0.20$.

Therefore, we conclude that formula (11) reproduces the simulation results of $P_L(r)$ and $P_{\text{link}}(r)$ for the cases of $r_d \leqslant 0.20$.

Let us give a remark. Generalizing (11), we introduce another fitting formula as follows. For $i = L$ or *link* we have

$$P_i(r) = \exp(-\alpha r^3) - C\exp(-\beta r^{\nu_1}) + D\exp(-\gamma r^3). \tag{12}$$

Here we remark that it has six fitting parameters.

Applying formula (12) to the same data points of $P_{\text{link}}(r, N, r_d)$, i.e. to the cases of $N = 32$, 64, 128 and 256 with the seven different values of excluded volume parameter r_d from $r_d = 0.0$ to 0.30, we have obtained good fitting curves to all the data including the case

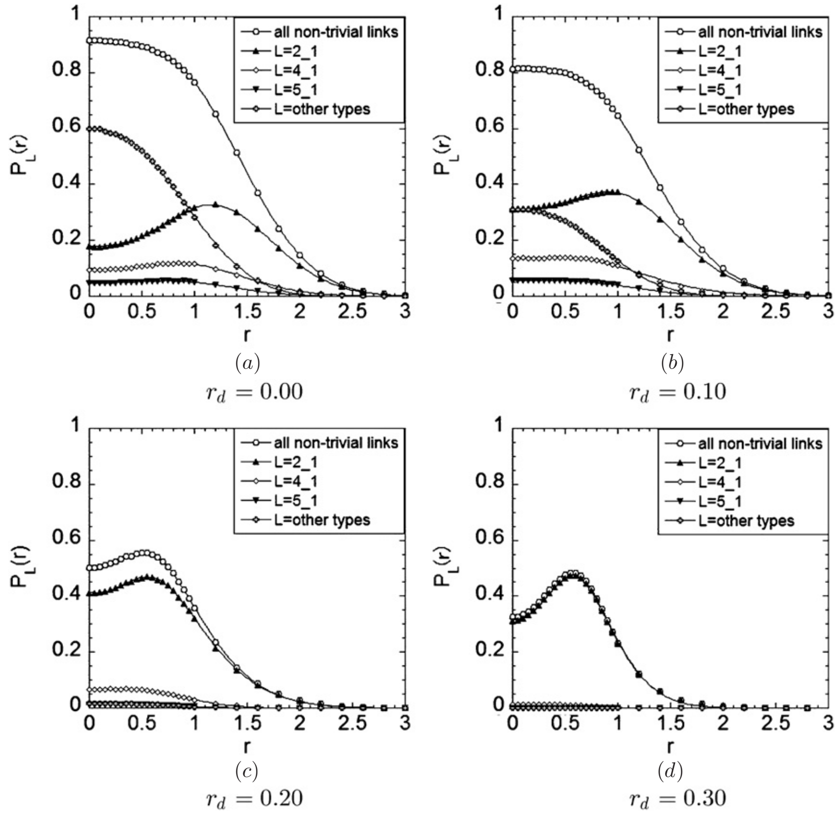


Figure 5. Linking probability $P_L(r, N, r_d)$ with link type L versus distance r for the SAP of nodal number $N = 256$ with the excluded volume $r_d = 0.0, 0.1, 0.2, 0.3$. Data from our simulation are represented by these points, where open circles are probabilities of nontrivial links P_{link} and closed triangles, open diamonds and closed inverted triangles are the probabilities P_{2_1}, P_{4_1}, P_{5_1}, respectively, and the open crosses are the probabilities P_{others} with more complicated link types, and solid lines are fitting curves by equation (11).

of large excluded volume such as $r_d = 0.25$ and 0.30. For all the 28 cases, χ^2 values are given by less than about 30. Thus, they are good also with respect to χ^2 values. Here we note that in the case of formula (11) the χ^2 values for $r_d = 0.25$ and 0.30 are given by larger than 150.

For an illustration, the best estimates of the six fitting parameters of formula (12) for $P_{\text{link}}(r, N, r_d)$ with $N = 256$ are given in table 4 for the seven values of the parameter r_d from $r_d = 0.0$ to 0.30.

4.2. Intuitive derivation of formula (11)

Let us discuss an intuitive derivation of the linking probability of $P_{\text{link}}(r)$ for $r_d = 0$, i.e. the probability of a given pair of random polygons separated by distance r between the centers of mass being equivalent to a nontrivial link [8]. The derivation is not rigorous but may give some hints to the reason why formula (11) gives good fitting curves.

Let us now discuss the large r-dependence of $P_{\text{link}}(r)$ for $r_d = 0$. We assume an ensemble of random polygons of N nodes.

(i) Choose a pair of N-noded polygons, randomly, from the ensemble. Place the two polygons in a way such that their centers of mass is of a distance r.

Table 2. Parameters of linking probability $P_{\rm link}(r)$ and the χ^2 values with respect to equation (11). N is the nodal number of a SAP and r_d is the size of the excluded volumes. The fitting curves of $P_{\rm link}(r)$ have five parameters denoted by α, ν_1, C, β and ν_2 of equation (11).

r_d	α	ν_1	C	β	ν_2	χ^2
			$N = 32$			
0.00	0.552 ± 0.008	2.319 ± 0.021	0.260 ± 0.001	1.595 ± 0.092	2.401 ± 0.054	3.719
0.05	0.568 ± 0.008	2.284 ± 0.021	0.267 ± 0.001	1.692 ± 0.094	2.354 ± 0.052	7.139
0.10	0.639 ± 0.008	2.192 ± 0.018	0.309 ± 0.001	1.983 ± 0.091	2.330 ± 0.044	8.670
0.15	0.790 ± 0.008	2.050 ± 0.017	0.396 ± 0.002	2.472 ± 0.095	2.197 ± 0.034	9.198
0.20	1.047 ± 0.007	1.879 ± 0.012	0.506 ± 0.002	3.757 ± 0.113	2.175 ± 0.028	26.76
			$N = 64$			
0.00	0.425 ± 0.005	2.476 ± 0.016	0.184 ± 0.001	1.665 ± 0.091	2.816 ± 0.079	7.502
0.05	0.440 ± 0.005	2.449 ± 0.016	0.195 ± 0.001	1.732 ± 0.091	2.772 ± 0.074	11.19
0.10	0.526 ± 0.006	2.320 ± 0.018	0.255 ± 0.001	1.831 ± 0.091	2.403 ± 0.054	7.616
1.50	0.735 ± 0.007	2.063 ± 0.015	0.369 ± 0.002	2.599 ± 0.097	2.239 ± 0.037	10.67
0.20	1.016 ± 0.006	1.870 ± 0.012	0.496 ± 0.002	3.935 ± 0.118	2.201 ± 0.029	27.62
			$N = 128$			
0.00	0.327 ± 0.003	2.667 ± 0.014	0.126 ± 0.001	1.828 ± 0.116	3.186 ± 0.116	25.63
0.05	0.344 ± 0.003	2.632 ± 0.015	0.139 ± 0.001	1.815 ± 0.108	3.085 ± 0.104	18.08
0.10	0.458 ± 0.005	2.400 ± 0.015	0.213 ± 0.001	2.059 ± 0.099	2.618 ± 0.065	6.301
0.15	0.693 ± 0.006	2.093 ± 0.014	0.350 ± 0.002	2.689 ± 0.100	2.274 ± 0.039	14.32
0.20	1.008 ± 0.006	1.870 ± 0.011	0.496 ± 0.002	4.061 ± 0.121	2.209 ± 0.029	43.41
			$N = 256$			
0.00	0.258 ± 0.002	2.870 ± 0.013	0.085 ± 0.001	2.257 ± 0.178	3.761 ± 0.172	43.42
0.05	0.277 ± 0.002	2.812 ± 0.013	0.098 ± 0.001	2.152 ± 0.154	3.498 ± 0.146	33.30
0.10	0.408 ± 0.004	2.491 ± 0.014	0.184 ± 0.001	2.206 ± 0.111	2.752 ± 0.075	12.96
0.15	0.678 ± 0.005	2.104 ± 0.013	0.340 ± 0.002	2.920 ± 0.109	2.309 ± 0.041	12.20
0.20	1.001 ± 0.005	1.859 ± 0.011	0.493 ± 0.002	4.289 ± 0.127	2.241 ± 0.029	51.30

(ii) If they are unlinked, they should be unlinked when they are placed with distance $r + \mathrm{d}r$ between the centers of mass. If they are linked, then they may become unlinked when they are placed with distance $r + \mathrm{d}r$ between the centers of mass.

(iii) The decrease $dP_{\rm link}(r)$ should be approximately proportional to the product of $P_{\rm link}(r)$ (being linked) and the partial volume $4\pi r^2\,\mathrm{d}r$ of the configuration space

$$\mathrm{d}P_{\rm link}(r) = -C_\ell P_{\rm link}(r) \times 4\pi r^2\,\mathrm{d}r. \tag{13}$$

(iv) Integrating the above differential equation we have

$$P_{\rm link}(r) = P_{\rm link}(0)\exp(-\alpha r^3),$$

where $\alpha = 4\pi C_\ell/3$.

If we assume that constant C_ℓ depends on r as $C_\ell(r) = C_0 r^{\nu_1 - 3}$, then by integration we have

$$P_{\rm link}(r) = P_{\rm link}(0)\exp(-\alpha r^{\nu_1}),$$

where $\alpha = 4\pi C_0/\nu_1$.

Table 3. Parameters of linking probability $P_{2_1}(r)$ with the link type 2_1 and the χ^2 values with respect to equation (11). N is the nodal number of a SAP and r_d is the size of the excluded volumes. The fitting curves of $P_{2_1}(r)$ have five parameters denoted by α, ν_1, C, β and ν_2 of equation (11).

r_d	α	ν_1	C	β	ν_2	χ^2
			$N = 32$			
0.00	0.590 ± 0.012	2.249 ± 0.027	0.598 ± 0.001	1.376 ± 0.045	2.330 ± 0.023	3.959
0.05	0.604 ± 0.012	2.222 ± 0.026	0.593 ± 0.002	1.408 ± 0.046	2.302 ± 0.023	10.07
0.10	0.669 ± 0.012	2.148 ± 0.024	0.572 ± 0.002	1.558 ± 0.051	2.270 ± 0.024	9.455
0.15	0.817 ± 0.011	2.017 ± 0.021	0.553 ± 0.002	1.998 ± 0.063	2.176 ± 0.023	13.26
0.20	1.075 ± 0.008	1.847 ± 0.014	0.577 ± 0.002	3.217 ± 0.089	2.096 ± 0.023	25.26
			$N = 64$			
0.00	0.483 ± 0.011	2.315 ± 0.028	0.669 ± 0.001	1.125 ± 0.032	2.439 ± 0.022	11.34
0.05	0.500 ± 0.010	2.322 ± 0.027	0.659 ± 0.001	1.162 ± 0.033	2.446 ± 0.022	9.855
0.10	0.572 ± 0.012	2.240 ± 0.027	0.609 ± 0.001	1.287 ± 0.042	2.342 ± 0.023	9.459
0.15	0.769 ± 0.012	2.021 ± 0.022	0.564 ± 0.002	1.827 ± 0.060	2.132 ± 0.023	10.89
0.20	1.066 ± 0.008	1.809 ± 0.013	0.576 ± 0.002	3.345 ± 0.090	2.108 ± 0.023	19.12
			$N = 128$			
0.00	0.419 ± 0.010	2.424 ± 0.030	0.749 ± 0.001	0.942 ± 0.025	2.533 ± 0.019	20.37
0.05	0.437 ± 0.010	2.398 ± 0.029	0.730 ± 0.001	0.976 ± 0.026	2.541 ± 0.020	15.10
0.10	0.537 ± 0.011	2.247 ± 0.026	0.649 ± 0.001	1.198 ± 0.034	2.409 ± 0.022	8.701
0.15	0.750 ± 0.011	2.019 ± 0.021	0.576 ± 0.002	1.771 ± 0.055	2.155 ± 0.022	21.22
0.20	1.070 ± 0.008	1.799 ± 0.013	0.583 ± 0.002	3.370 ± 0.089	2.104 ± 0.022	33.89
			$N = 256$			
0.00	0.383 ± 0.010	2.484 ± 0.032	0.821 ± 0.001	0.881 ± 0.021	2.626 ± 0.017	16.73
0.05	0.399 ± 0.011	2.457 ± 0.032	0.798 ± 0.001	0.847 ± 0.022	2.598 ± 0.017	17.40
0.10	0.518 ± 0.011	2.262 ± 0.026	0.685 ± 0.001	1.108 ± 0.030	2.475 ± 0.021	10.09
0.15	0.766 ± 0.011	1.984 ± 0.020	0.587 ± 0.002	1.836 ± 0.054	2.154 ± 0.021	13.41
0.20	1.083 ± 0.007	1.773 ± 0.013	0.586 ± 0.002	3.487 ± 0.090	2.101 ± 0.022	37.11

Table 4. Six fitting parameters of $P_{\rm link}(r)$ and the χ^2 values with formula (12).

r_d	α	C	β	ν	D	γ	χ^2
				$N = 256$			
0.00	0.55 ± 0.05	0.75 ± 0.05	0.75 ± 0.04	3.08 ± 0.03	0.67 ± 0.05	0.20 ± 0.01	12.37
0.05	0.56 ± 0.05	0.74 ± 0.05	0.76 ± 0.05	3.06 ± 0.03	0.64 ± 0.05	0.20 ± 0.01	9.13
0.10	0.61 ± 0.05	0.67 ± 0.04	0.84 ± 0.07	2.93 ± 0.05	0.48 ± 0.04	0.21 ± 0.01	6.92
0.15	0.75 ± 0.09	0.69 ± 0.02	0.99 ± 0.18	2.57 ± 0.11	0.35 ± 0.02	0.23 ± 0.01	14.96
0.20	1.12 ± 0.07	0.73 ± 0.01	1.60 ± 0.15	2.41 ± 0.08	0.24 ± 0.01	0.26 ± 0.01	16.86
0.25	1.44 ± 0.05	0.77 ± 0.01	2.33 ± 0.10	2.34 ± 0.05	0.15 ± 0.01	0.30 ± 0.01	20.45
0.30	1.73 ± 0.04	0.81 ± 0.01	3.09 ± 0.09	2.45 ± 0.03	0.14 ± 0.01	0.37 ± 0.01	30.02

The above intuitive argument gives the first term of formula (11). We can derive the second term if we consider contributions from other link conditions in the right-hand side of (13). The second terms can be considered as a correction to the first term of (11).

We find in table 2 that the fitting parameter C of formula (11) for $P_{\text{link}}(r)$ becomes very small for random polygons with $r_d = 0$ and large N. Furthermore, ν_1 becomes close to 3.0 as N increases. Thus, the above intuitive argument should be appropriate for random polygons of very large N and small r_d.

By a similar intuitive argument we can derive formula (11) also for other link types. For instance, in the Hopf link case, we obtain the difference of two exponentials as follows [25]:

$$P_{2_1}(r) = B_1 \exp(-\beta_1 r^3) - B_2 \exp(-\beta_2 r^3). \tag{14}$$

Let us recall the derivation of formula (14), [25]. We first assume the following:

(i) If a given pair of polygons with distance r between the centers of mass gives the trivial link, then it should also be trivial when the distance between the centers of mass is given by $r + \mathrm{d}r$.
(ii) If a given pair of polygons with distance r between the centers of mass gives the Hopf link, then it may become a different link when the distance between the centers of mass is given by $r + \mathrm{d}r$.
(iii) If the pair is neither the trivial nor the Hopf link, it may become a Hopf link when the distance between the centers of mass is given by $r + \mathrm{d}r$.

Then we have

$$\mathrm{d}P_{2_1}(r) = -\gamma_1 P_{2_1}(r)\,\mathrm{d}v + \gamma_2(P_{\text{link}}(r) - P_{2_1}(r))\,\mathrm{d}v, \tag{15}$$

where $v = 4\pi r^3/3$, and γ_1 and γ_2 are constants. By integrating (15) we have (14).

Similarly, if we assume a sequence of links from simple to complex links, through similar intuitive arguments, the linking probability can be expressed as a sum of several exponential terms of r^3.

5. Dependence of the linking probabilities on the excluded volume r_d and the number of segments N

5.1. Dependence of linking probability on the excluded volume

Let us now discuss the r_d-dependence and the N-dependence of the linking probabilities P_L and P_{link} which we have investigated for the simulation data.

We have found several features of the r_d-dependence of $P_{\text{link}}(r)$. The estimates of $P_{\text{link}}(r)$ of SAP containing 256 nodes are shown against distance r for various values of r_d in figure 6. We have the following observations:

(i) The number of nontrivial links decreases as the excluded volume parameter r_d increases.
(ii) For $r_d \geqslant 0.20$, a peak appears in the graph of $P_{\text{link}}(r)$ as a function of distance r.
(iii) The height and the position of the peak does not change for $r_d \geqslant 0.25$.
(iv) The probability $P_{\text{link}}(r)$ does not vanish even for $r_d = 0.30$.

Let us explain observation (i) as follows. The effective repulsions among the segments of SAP arising from the excluded volume become stronger as parameter r_d increases. The segments of two SAPs are effectively repelled by each other so that the range of the distribution function of the SAPs should be extended and the local density of segments of SAPs should decrease. Thus, the degree of entanglement between the two SAPs should also decrease with respect to parameter r_d, and the linking probability should become smaller. Here we have assumed that the degree of entanglement between two SAPs should be proportional to the local density of segments of the SAPs.

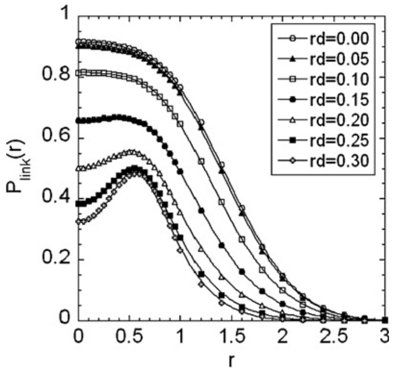


Figure 6. Linking probability $P_{\text{link}}(r, N, r_d)$ versus distance *r* for SAPs of $N = 256$ with the following seven values of the excluded volume parameter r_d: $r_d = 0.0$ (open circles); 0.05 (closed triangles); 0.10 (open squares); 0.15 (closed circles); 0.20 (open triangles); 0.25 (closed squares); 0.30 (open crosses). Solid lines are fitting curves given by formula (11).

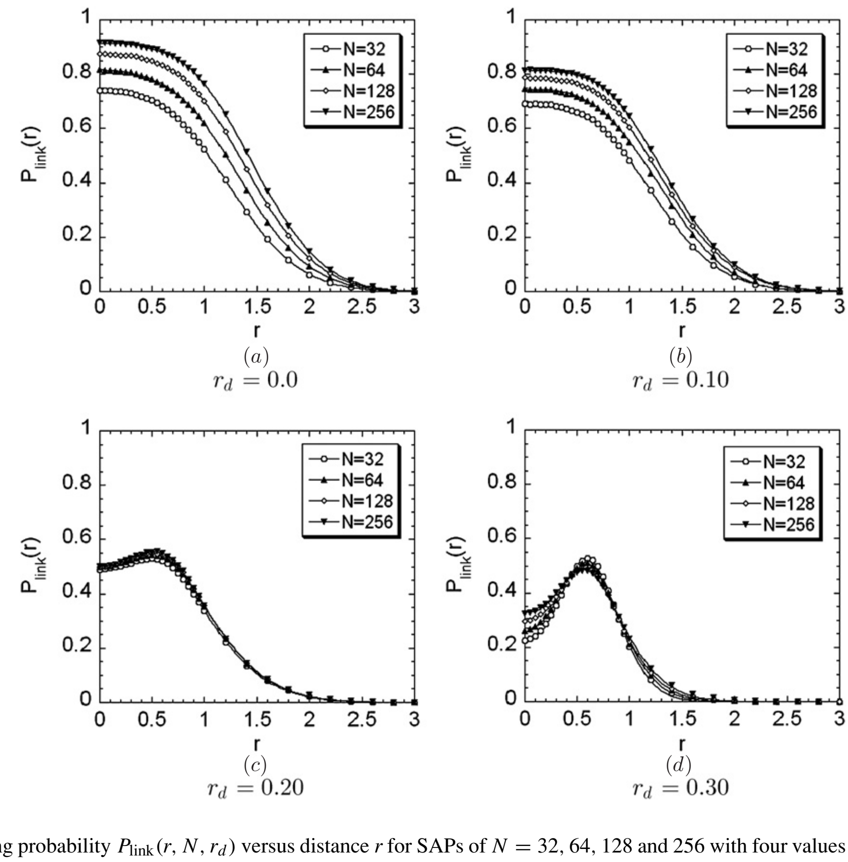
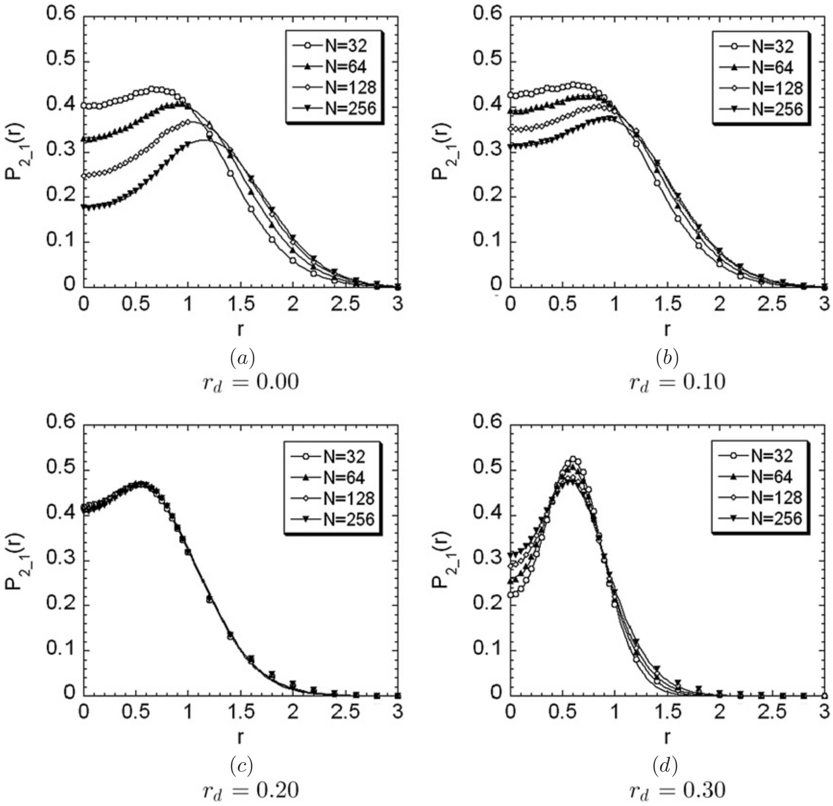


Figure 7. Linking probability $P_{\text{link}}(r, N, r_d)$ versus distance *r* for SAPs of $N = 32$, 64, 128 and 256 with four values of the excluded volume parameter: (a) $r_d = 0.0$; (b) $r_d = 0.1$; (c)$r_d = 0.2$; (d) $r_d = 0.3$. Simulation data of the probability for $N = 32$, 64, 128 and 256 are represented by open circles, closed triangles, open diamonds and closed inverted triangles, respectively. Solid lines are given by formula (11).

We remark that observation (i) is also consistent with the fact that the probability for SAP to be a nontrivial knot becomes smaller for off-lattice SAP when the excluded volume parameter r_d increases [20].



(a) $r_d = 0.00$ (b) $r_d = 0.10$ (c) $r_d = 0.20$ (d) $r_d = 0.30$

Figure 8. Linking probability of link type 2_1 (P_{2_1}) versus distance *r* for the SAP of $N = 32, 64, 128$ and 256 with four values of the excluded volume parameter: (*a*) $r_d = 0.0$; (*b*) $r_d = 0.1$; (*c*) $r_d = 0.2$; (*d*) 0.3. Data points of the probabilities for $N = 32, 64, 128$ and 256, are represented by open circles, closed triangles, open diamonds and closed inverted triangles, respectively. Solid lines are given by formula (11).

We can explain observation (ii) from the behavior of the linking probability of some link types (P_L) as shown in figure 5. Here we find that for $r_d \geqslant 0.2$ the most nontrivial link is given by the Hopf link, 2_1. It should be difficult to generate complicated links when the effective repulsive force of the excluded volume acts on the segments of SAPs strongly. Therefore, the profile of $P_{\text{link}}(r)$ with respect to distance *r* is almost the same as that of $P_{2_1}(r)$. We remark that the graph of $P_{2_1}(r)$ has a peak, in the same way as the graph of $P_{\text{link}}(r)$ has a peak for $r_d \geqslant 0.2$.

It is suggested from observation (iii) and figure 5 that the fraction of the Hopf link (2_1) should be nonzero even if r_d reaches its limit: $r_d \to 0.5$.

5.2. *N-dependence of the linking probabilities*

Let us discuss the *N*-dependence of the linking probabilities, $P_{\text{link}}(r)$ and $P_L(r)$. The linking probability $P_{\text{link}}(r)$ and $P_{2_1}(r)$ for various *N* is plotted against distance *r* in figures 7 and 8, respectively.

One of the most important features of figure 7 is that when $r_d = 0.2$ probability $P_{\text{link}}(r)$ has the same value for all values of *N*. The *N*-independence of $P_{\text{link}}(r)$ for $r_d = 0.2$ is quite remarkable. Moreover, in figure 8 the linking probability of the Hopf link, $P_{2_1}(r)$, has also the

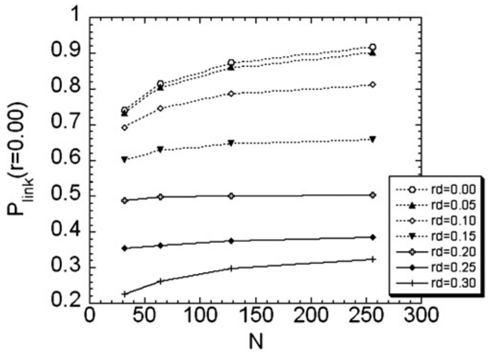


Figure 9. Linking probability $P_{\rm link}(r = 0.00)$ versus the nodal number *N*. The distance *r* between SAP is fixed as $r = 0.00$. These points represent our simulation results. Where the excluded volume $r = 0.00$ (open circles), 0.05 (closed triangles), 0.10 (open diamonds), 0.15 (closed inverted triangles), 0.20 (open crosses), 0.25 (closed diamonds) and 0.30 (+s).

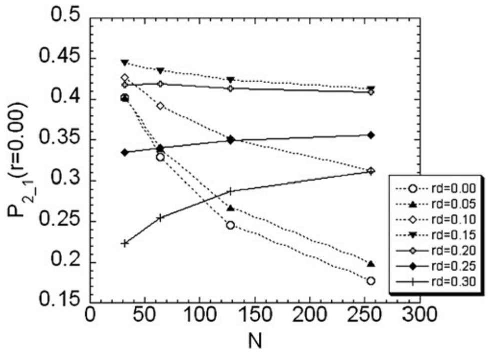


Figure 10. Linking probability $P_{2_1}(r = 0.00)$ versus the nodal number *N*. The distance *r* between SAP is fixed as $r = 0.00$. These points represent our simulation results. Where the excluded volume $r_d = 0.00$ (open circles), 0.05 (closed triangles), 0.10 (open diamonds), 0.15 (closed inverted triangles), 0.20 (open crosses), 0.25 (closed diamonds) and 0.30 (+s).

same *N*-independence for $r_d = 0.2$. In fact, we find that $P_L(r)$ for all other links *L* investigated show the same *N*-independence for $r_d = 0.2$.

In the case of $r_d < 0.20$, the graph of $P_{\rm link}(r)$ strongly depends on the number of segments, *N*. In fact, $P_{\rm link}(r)$ increases as *N* increases. For $r_d = 0.30$ the *N*-dependence of $P_{\rm link}(r)$ is weak but not zero. However, it is much weaker than that of $r_d < 0.2$. The *N*-dependence of the linking probability can be explained by the hypothesis that the longer are the SAP the more likely they entangle with each other.

Let us now discuss the *N*-dependence of $P_{\rm link}(r = 0.00)$. It is the probability when both the two centers of mass of SAPs are located at the origin. In figure 9 the estimates of $P_{\rm link}(r = 0.00)$ are plotted against the number of segments *N* for seven values of r_d such as $r_d = 0.00, 0.05, 0.10, 0.15, 0.20, 0.25$ and 0.30, respectively. Interestingly we find that $P_{\rm link}(r = 0.00)$ increases with respect to *N* for $r_d \neq 0.2$, but not for $r_d = 0.2$. The probability $P_{\rm link}(r = 0.00)$ does not depend on *N* for the case of $r_d = 0.2$.

Furthermore, let us discuss the *N*-dependence of $P_{2_1}(r = 0.00)$, the linking probability of the Hopf link at zero distance between the centers of mass of SAPs. The estimates of $P_{2_1}(r = 0.00)$ are shown against *N* in figure 10. Here we have three types of the *N*-

dependence: for $r_d < 0.20$, the probability $P_{2_1}(r = 0.00)$ decreases as N increases. This behavior is different from that of $P_{\text{link}}(r = 0.00)$. For $r_d = 0.20$, the probability $P_{2_1}(r = 0.00)$ does not depend on N. For $r_d > 0.2$, the probability $P_{2_1}(r = 0.00)$ increases with respect to N.

6. Conclusions

Through simulation using topological invariants of knots and links we have numerically evaluated the linking probability for a given pair of SAPs for various link types. In the simulation, we have assumed that every pair of SAPs should have no overlaps among spherical segments of radius r_d and each of the SAPs should have the trivial knot type.

More precisely, we define the linking probability of a link type L consisting of two trivial knots by the probability that the topology of a given self-avoiding pair of SAPs is equivalent to link type L. We have evaluated the linking probability of link type L as a function of distance r between the centers of mass of the two SAPs of N segments.

We have introduced two formulae expressing the linking probability as a function of distance r. They have five and six parameters, respectively. Both of them give good fitting curves with respect to χ^2 values.

We have also investigated the dependence of linking probabilities on the excluded volume parameter r_d and the number of segments, N. Quite interestingly, the graph of linking probability versus distance r shows no N-dependence at a particular value of the excluded volume, $r_d = 0.2$.

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