The knot problem in statistical mechanics of polymer chains

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The problem of accounting for topological restrictions in calculating the statistical integral for a closed polymer chain is treated. The topological invariants of knots are discussed. It is shown that the integral invariant proposed by Edwards is absolutely incorrect. The Monte Carlo method is used to calculate the probability of knot formation when a polymer chain is closed, and the results of the algebraic theory of knots (Alexander polynomials) are used to determine the type of knot. Calculations are carried out for two random-walk models on a lattice for chains of length L from 10 to 150 segments. The first model, which corresponds to a chain having a width close to the length of a segment, gives a low probability of knot formation ($\lesssim 10^{-3}$). The second model corresponds to the case of an infinitesimally thin chain, and it gives a much greater probability of knot formation, which increases approximately linearly in the studied range of lengths, and which attains a value of ≈ 0.4 for $L \approx 150$. Data were also obtained here on the frequency of formation of different types of knots. The possible role of the obtained results in the statistical mechanics of polymers and biopolymers is briefly discussed.

INTRODUCTION

An important unsolved problem of the statistical mechanics of polymer chains is how to account for topological restrictions in calculating the statistical integral for closed chains. This problem was first formulated clearly by Delbrück.[1] Edwards[2] has tried to devise a consistent statistical theory of closed polymer chains with account taken of topological restrictions. In devising a theory of this type, one must first of all possess an invariant of a knot, i.e., an algorithm that answers whether two closed chains are topologically equivalent or not. Edwards[2] has proposed an integral invariant of a knot. However, as we shall show below, Edwards' invariant is absolutely incorrect.

In line with this, we have turned to the algebraic (group-theoretical) approach. In contrast to the integral invariant, a knowledge of which might open up possibilities of analytical solution of the problem such as those that Edwards[2] tried to realize, the algebraic invariant can be used only by applying numerical methods. By using this invariant in the current study, we have performed computer calculations, by the Monte Carlo method, of the probability of formation of different types of knots as a chain is generated on a lattice.

THE INTEGRAL INVARIANT OF EDWARDS

In order to derive a topological integral invariant of a knot, Edwards[2] reduces this problem to that of a topological integral invariant for the linking of two closed chains, which has been treated by Gauss. The invariant of Gauss has the form

$$\oint\oint \frac{[d\mathbf{r}_1\, d\mathbf{r}_2]\mathbf{r}_{12}}{r_{12}^3}, \qquad (1)$$

where the integration is performed over the contours C_1 and C_2 (see Fig. 1a); $\mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2$. The integral (1) vanishes if the chains 1 and 2 are not linked, and is equal to $\pm 4\pi n$ for n-fold linking of the chains.[1)]

Edwards considers the same expression (1) for a single closed chain. However, he integrates both times over the same trajectory. In order to prove that this

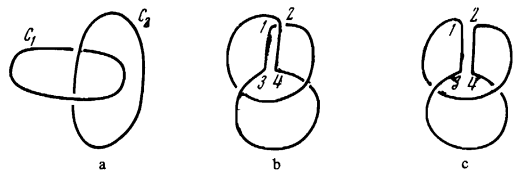

FIG. 1

expression can be used as a topological invariant for a chain containing knots, he presents the diagram that we have reproduced as Fig. 1b. The diagram shows a chain forming a knot. Since the integration over the segments 1–3 and 2–4 is performed in opposite directions, the integrals over these segments mutually cancel as they approach one another. Hence, the integral over the entire trajectory equals the integral over the two linked rings that are obtained after these segments have been merged and discarded. Hence, it differs from zero. However, a chain that does not contain a knot can be deformed into a plane ring for which the integral (1) evidently vanishes. The invariant introduced by this method is then used in[2] to calculate the statistical integral of a closed chain containing knots.

However, we can easily see that Edwards' treatment leads to a paradox. In fact, let us examine the chain depicted in Fig. 1c. On the one hand, the arguments given above for the chain of Fig. 1b that reduce it to two linked rings are fully applicable to it. On the other hand, it is not a knot, and it can be deformed into a plane ring. We see that Edwards' invariant has an evident flaw.

Edwards' arguments assume that the value of the integral (1) taken over a single closed chain does not vary under all possible deformations without self-intersection. Naturally, any topological invariant must possess this property. Yet the value of the integral (1) for a single closed chain depends substantially on its concrete form.

To give a pictorial demonstration of this fact, we have calculated the value of the integral (1) for different chains arranged on a cubic lattice. In this case, the integration by the formula (1) can be completely per-

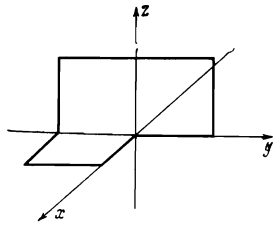

FIG. 2

formed, and for example, the value of the integral for the contour shown in Fig. 2 proves to be the following:

$$12\operatorname{arctg}\frac{1}{\sqrt{3}}-4\operatorname{arctg}\frac{2}{\sqrt{6}}-2\operatorname{arctg}\frac{1}{2\sqrt{6}}=\pi.$$

Evidently, a trajectory having a center or plane of symmetry makes the integral (1) vanish. The figure shown in Fig. 2 is a very simple asymmetric contour on a cubic lattice, and for it the integral (1) no longer vanishes. Hence there are no grounds for thinking that the integral (1) taken over a single closed chain must vanish for any conformations of the chain except those that have a center or plane of symmetry. Thus, the integral proposed by Edwards is not a topological invariant at all—its value changes upon simple deformations of the contour not involving a change of topology.

THE ALGEBRAIC INVARIANT OF A KNOT

Knots as mathematical objects first attracted attention in the sixties of the past century. The origin of the theory of knots is associated with the name of the English physicist and mathematician Tait. (see[3,4]). The modern theory of knots is one of the branches of algebraic topology. An excellent presentation of the fundamental concepts of the theory, as well as a long list of the most important studies in this field can be found in the monograph of Crowell and Fox.[5]

One of the major attainments of the theory of knots was the creation and rigorous justification of the algebraic invariant of a knot. Without entering into the mathematical details, which can be found in[5], we shall present in this section the process of calculating this invariant in a form that can be easily programmed for computer calculations.

First let us introduce a set of definitions (see[5]). Two closed contours are equivalent knots if they can be transformed into one another by continuous deformation of the contour without self-intersections.[2)] For convenience of further discussion, the knot is deformed into a polygonal form, in which it is a sequence of a finite number of linear segments called sides. The ends of these sides are called the vertices. One can represent a knot by its projection on some plane, which we shall denote as the xy plane. We shall call a point of this plane that contains the projections of more than one point of the knot a multiple point. We choose a projection of the knot that puts it into a regular position in which: 1) all of its multiple points are double, and there is a finite number of them; 2) no double point is the image of a vertex of the knot. Then each double point corresponds to an intersection. The point having the smaller z coordinate is called the undercrossing, and the other, respectively, the overcrossing. Figure 3 shows the regular positions of two knots. The knot a is called a trefoil, and knot b, as we can easily see, is trivial (it is equivalent to a circle).

It will be convenient henceforth to distinguish an arbitrary point 0 that is not multiple on the contour of a knot in a regular position, and to assign (also arbitrarily) the direction of passage of the contour of the knot. Now we shall move along the direction of passage of the contour. Let the point where we are at a given moment for some intersection be the undercrossing point. We shall call this intersection an underpass, and the other case an overpass. As we pass around the contour, we meet each intersection twice, once as underpass, and the other time, as overpass. Now let us number all the underpasses, and we shall consider underpass No. 1 to be the first underpass that we met in moving along the contour from point 0 (see Fig. 3). Evidently, we have numbered all the intersections existing in a given regular position. The part of the contour lying between the (k − 1)st and k-th underpasses is called the k-th generator, and is denoted by x_k. The generator $x_1 = x_{n+1}$ (n is the total number of intersections) lies between the first and last underpasses. We shall always denote it as x_1 (see Fig. 3). The underpasses can be of two types (I and II), depending on the direction of the overpassing generator (see Fig. 4). Thus, each regular position of a knot is characterized by a sequence of underpasses, for each of which its type (I and II) and the number of the overpassing generator is specified.

One can represent such a description of a regular position as a square matrix, which is called the Alexander matrix. The kth row of this matrix corresponds to the kth underpass, and it consists of n elements a_{kj} (j = 1, . . . , n; n is the number of underpasses.) Almost all of these elements are equal to zero; only the elements a_{kk}, $a_{k,k+1}$ and a_{ki} differ from zero. The latter are equal to[3)] (i is the number of the overpassing generator of the kth underpass):

1) when i = k or i = k + 1, independently of the type of underpass,

$$a_{kk}=-1,\quad a_{k\,k+1}=1,$$

2) when i ≠ k, i ≠ k + 1 for a type I underpass,

$$a_{kk}=1,\quad a_{k\,k+1}=-t\quad a_{ki}=t-1$$

and for a type II underpass,

$$a_{kk}=-t,\quad a_{k\,k+1}=1,\quad a_{ki}=t-1.$$

The Alexander polynomial Δ(t), which is an invariant of the knot, is derived from the Alexander matrix as

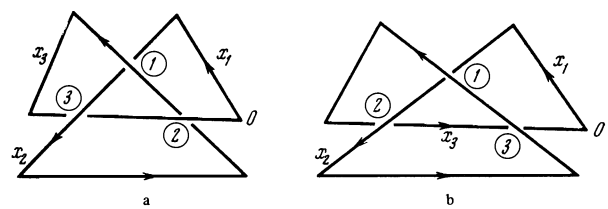

FIG. 3

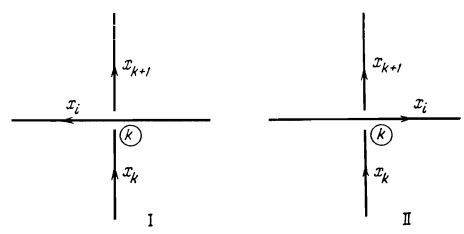

FIG. 4

follows. One calculates any minor of order n − 1 (the result does not depend on the choice of minor), and multiplies it by the quantity $\pm t^{-m}$ (m is an integer) in such a way that the obtained polynomial does not contain negative powers, and has a positive free term.

It is rigorously proved in knot theory that the Alexander polynomials $\Delta(t)$ coincide for equivalent knots. Thus, for a trivial knot $\Delta(t) = 1$, for a trefoil $\Delta(t) = t^2 - t + 1$, etc (see[5-7]). We should note that the above-cited correspondence between the rows of the Alexander matrix and the two types of underpasses (I and II) is arbitrary. The only important point is that the given assignment should remain fixed as the matrix is compiled for the whole knot. The interchange I ⟷ II for all the underpasses does not alter the Alexander polynomial $\Delta(t)$. Yet such an operation corresponds to a mirror reflection of the knot. Hence, the given invariant does not permit one to distinguish a knot and its mirror image, although in many cases these knots prove to be topologically nonequivalent (this problem has been discussed in the monograph[5]).

In the mathematical literature, knots are generally classified according to the minimum number of intersections in their projections (see[3,4,6,7]). Knots having the same number of intersections are arranged in some definite sequence, and the corresponding order number is added as a subscript to the number of intersections. Figure 5 gives the four simplest nontrivial knots and the corresponding Alexander polynomials.[4)] This type of classification has been carried out for all 84 knots having a minimum number of intersections less than 10, and the Alexander polynomials have been calculated for them (see[6,7]). This classification pertains to the so-called simple knots. In addition, various combinations can be formed among them (composite knots). Here the Alexander polynomial of the composite knot equals the product of the polynomials of its constituent knots.

We should note that the Alexander polynomial is not a complete invariant of a knot. There are examples of nonequivalent knots that have identical Alexander polynomials. Thus, $\Delta(t) = 2t^2 - 5t + 2$, both for the knot 6_1 and for 9_{46}. However, such cases happen rather rarely. For the 84 simple knots mentioned above, the Alexander polynomials coincide for five pairs (see[6,7]). An essential point here is that $\Delta(t) \pm 1$ for all 84 of these knots. That is, this invariant distinguishes them from the trivial knot. Nevertheless, there are nontrivial knots whose Alexander polynomials equal unity.[8] However, they are very complex. The simplest knot of this type (Fig. 6) contains 11 intersections.

Thus, the Alexander polynomial is an invariant of a knot that is most suitable for the purposes of this study. On the one hand, the algorithm for calculating it is rather easy, and it can easily be programmed on a computer. On the other hand, this invariant appears to be good enough, and as a rule, it permits one not only to distinguish trivial from nontrivial knots, but also to distinguish nontrivial knots of different types.

CALCULATIONS OF PROBABILITIES OF KNOT FORMATION

The fundamental problem of the statistical mechanics of knots consists in determining what fraction of all possible closed trajectories consisting of a given number of segments belongs to a given topological class. In particular, it is of interest to find out what fraction of

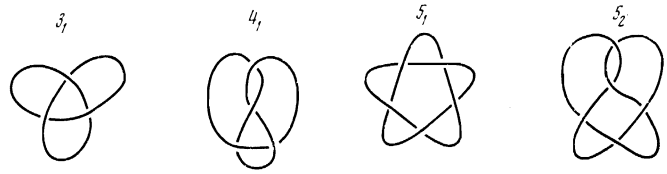

FIG. 5. The simplest nontrivial knots; they correspond to the following Alexander polynomials: knot 3_1–polynomial $\Delta(t) = t^2 - t + 1$; knot 4_1 – $\Delta(t) = t^2 - 3t + 1$; knot 5_1 – $\Delta(t) = t^4 - t^3 + t^2 - t + 1$; knot 5_2 – $\Delta(t) = 2t^2 - 3t + 2$.

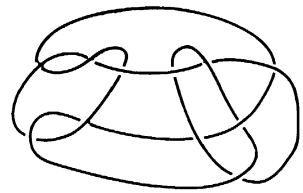

FIG. 6. The simplest nontrivial knot for which $\Delta(t) = 1$.

these trajectories forms a trivial knot in the range of lengths that is typical of actually encountered polymer chains. We have used the Monte Carlo method to answer these questions. The calculations were run on the BÉSM-6 computer.

The trajectories on a three-dimensional lattice were obtained by modeling with a random walk without self-intersection, Only the closed trajectories were considered among all those having a given length L (more exactly, those having a distance between the ends of unity, since they can be closed without ambiguity). We used a special method, which is described in detail in the Appendix, to increase the fraction of closed trajectories, We analyzed each closed trajectory by using the algorithm presented in the last section. In practice, we calculated the Alexander polynomial $\Delta(t)$ for distinguishing knots of different types only for two values of the argument: $t = -1$ and $t = -2$. Analysis of the table of Alexander polynomials for the 84 types of simple knots[6,7] shows that a knowledge of the values $\Delta(-1)$ and $\Delta(-2)$ permits one unambiguously to establish which Alexander polynomial they correspond to.

The calculations of probabilities of knot formation were performed for two random-walk models. In the first model, the random walk without self-intersection was directly carried out on a body-centered lattice. It turned out that the probability of formation of nontrivial knots in this model is very small in the region of chain lengths that we studied ($\sim 10^{-5}$–10^{-3}). Here practically all of the obtained nontrivial knots belonged to the type 3_1 (trefoil). Such a small probability of knot formation might involve the fact that the studied model actually corresponds to a "thick" polymer chain for which the thickness of a segment is close to its length. In fact, in a random walk without self-intersection on a lattice, remote segments along the chain cannot approach one another to a distance less than the lattice period.

In line with this, a second model was studied that corresponds to a polymer chain consisting of infinitesimally thin segments. In this model the random walk was carried out as follows. Up to the first intersection, the random walk was carried out in the usual way with backward steps forbidden. When the end of a segment fell at an already occupied lattice mode, a new lattice was introduced, just like the original one, but shifted with respect to it along some randomly chosen direction by a small amount (as compared with the lattice period).

The random walk was continued along this new lattice until the next intersection, whereupon the next lattice was introduced, again shifted with respect to the previous one by a small amount. This procedure permits one to get trajectories that contain no self-intersections, but in which segments that are remote along the chain can approach one another to a very short distance. It turned out that the probability of knot formation is sharply increased in this model. Moreover, in this model one gets a great variety of knots of different types. The results of the calculations are given in the table and in Fig. 7

First of all we see that the probability of formation of a nontrivial knot in the studied length range increases almost linearly with increasing number L of segments, and it attains a value ≈ 0.4 at $L \approx 150$, without exhibiting yet any appreciable tendency to saturation. The frequency of occurrence of a knot of a given type among the knotted chains declines rapidly with increasing complication of the knot (increasing number of intersections). Here one finds knots having more than nine intersections extremely rarely in the studied range of lengths (see the last column of the table). This indicates that the small uncertainty that can arise in using the Alexander polynomial as an invariant of a knot (see the last section) plays practically no role in our calculations.

DISCUSSION OF RESULTS

Thus we have found the probability of knot formation upon closing chains consisting of a number of segments lying in the range from 10 to 150. This probability has proved to depend substantially, not only on the number of segments in the chain, but also on the thickness of a segment (more exactly, on the relationship between the thickness and length). It would be highly desirable, of course, to proceed to greater chain lengths. This is limited both by the memory and the speed of the computer that we have used.

However, since there have been no estimates of this type at all up to now, the results already obtained can be of interest in treating an entire set of problems.

Let us list some of them.

1. The problem was discussed in[9] of the possible role of knots in the theory of the helix-coil transitions in DNA, we can conclude from the results obtained in the present study that accounting for knots need not lead to substantial corrections in the study of heteropolynucleotides, but must be extremely significant in studying melting of homopolynucleotides. In particular, the theoretical prediction of the possibility of phase transitions in the homopolymers must be reexamined. However, we cannot do this as long as we lack an asymptotic estimate of the fraction of knotted trajectories.

2. The results obtained for the case of an infinitesimally thin chain can be directly applied for estimating the probability of knot formation when DNA is closed into a ring. This involves the fact that the thickness of two-stranded DNA (20 Å) is very small in comparison with the length of the segment ($\approx$1000 Å). In particular, we can conclude that when the DNA of λ phage, which contains about 150 segments, is closed, about half of the molecules must be knotted. Experiments on cyclization of λ phage DNA in solution have been performed repeatedly (see[10]). However, the molecules obtained here have not been analyzed, insofar as we know, for formation of nontrivial knots in them.

3. The problem of knots in polymers has been repeatedly discussed also in the chemical literature (see, e.g.,[11-13]). One of the ways of synthesizing knotted molecules that has been discussed in the literature consists in trying to select from a mixture obtained by random closing of chains. The results of these calculations show that this method in highly ineffective for the case of flexible (i.e., "thick") chains. This apparently is involved in the failures in attempts to synthesize knots based on hydrocarbon chains by the method of random closing.

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Results of calculations of probabilities of formation of nontrivial knots of different types for the second model (the "thin" chain)

Length of trajectory	Number of trajectories	Number of knots	Probability of knot formation, %	Percent of knots of different types					
				3_1	4_1	5_1	5_2	from 6 to 9	greater than 9
8	23500	0	0	—	—	—	—	—	—
10	15180	38	0.25±0.05	98	2	0	0	0	0
12	21380	132	0.6±0.05	95	4.5	0	0.5	0	0
16	9700	113	1.2±0.1	85	11	1	1	2	0
20	12560	293	2.3±0.1	83	10	0.5	2.5	3	0
30	5950	289	4.9±0.3	77	13	1	5	3.5	0.5
40	3000	240	8.0±0.6	76	12	5	3	4	0
50	2500	270	10.8±0.6	67	16	3.5	7.5	5.5	0.5
60	3450	481	14.0±0.6	66	13	4	6	7.5	0.5
70	1460	265	18.1±1.0	60	12	3	7	16.5	1.5
80	1340	275	20.5±1.0	58	13	6	6	15	2
90	1120	253	22.7±1.2	62	13.5	2.5	8.5	12.5	1
100	910	227	25.2±1.4	58	14.5	2.5	6.5	16	2.5
110	400	116	29.2±2.3	50	13.5	2	5	25	4.5
120	415	147	35.5±2.4	58.5	9	1.5	6	20	5
130	520	168	32.3±2.0	55	9	2.5	11	20.5	2
140	430	157	36.5±2.3	53.5	7.5	1.5	9.5	23	5

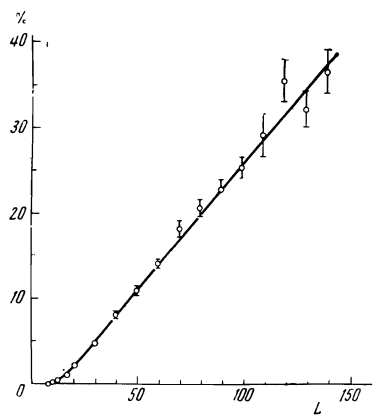

FIG. 7. Probability of formation of a nontrivial knot by a closed polymer chain consisting of L segments for the second model (the "thin" chain).

APPENDIX

GENERATION OF CLOSED CHAINS ON A LATTICE

It is highly ineffective to generate all possible chains and select the closed ones from them because the probability of closure of a trajectory of a given length in a random walk is very small. In order to hasten the closure process, we assumed the probabilities of different directions of the i + 1-st step not to be equal, but to be given by a distribution function $P(L - i, r_i, \delta)$, where r_i is the radius vector of the end of the ith link of the chain, and δ is a vector characterizing the direction of the i + 1-th link. The quantity $P(L - i, r_i, \delta)$ is a provisional distribution function that takes account of the fact that the trajectory must become closed at the L-th

step. We can easily find the function $P(L-i, r_i, \delta)$ for a body-centered lattice, neglecting the self-intersection of the chain. This involves the fact that random walks on a body-centered lattice can be reduced to three independent one-dimensional random walks over the coordinates x_1, x_2, and x_3 with a displacement of ± 1 at each step. Consequently, the distribution function of the coordinates of the end of the trajectory at the k-th step as referred to the origin of the random walk is

$$P(k,x)=\frac{1}{8^k}\prod_{i=1}^{3}C_k^{(k-x_i)/2}, \qquad (A.1)$$

In order to calculate the distribution function $P(L-i, r_i, \delta)$, let us examine the remaining part of the closed trajectory of length $L-i-1$ that joins the point of radius vector $r_i+\delta$ with the origin, with δ adopting a sequence of values. The sought distribution function is the ratio of the number of trajectories that correspond to a given value of δ to the total number of trajectories that correspond to all possible values of δ, i.e.,

$$P(L-i,\ r_i,\delta)=P(L-i-1,\ r_i+\delta)\Big/\sum_{\delta}P(L-i-1,r_i+\delta).$$

According to the Chapman-Kolmogorov equation,

$$P(L-i,r_i)=\frac{1}{l}\sum_{\delta}P(L-i-1,r_i+\delta),$$

where l is the coordination number of the lattice. Hence,

$$P(L-i,r_i,\delta)=P(L-i-1,r_i+\delta)/lP(L-i,r_i). \qquad (A.2)$$

Equations (A.1) and (A.2) imply that, for the studied body-centered lattice,

$$P(n,r,\delta)=\prod_{k=1}^{3}\frac{n-\delta_k x_k}{2n}, \qquad (A.3)$$

where $\delta_k=\pm 1$.

The described method for increasing the fraction of closed chains is not fully adequate because the function (A.3) does not take account of the rule against self-intersection of the chain. Actually a rule against self-intersection was introduced in constructing the closed chains. That is, the chain can be extended only to unoccupied lattice nodes (in the case of the "thin" filament, this corresponded to a rule against backward steps). In calculating the means by the Monte Carlo method, one must ascribe in this case a definite weight to each obtained trajectory (see[14,15]). This weighting factor was calculated as the product (over all the steps) of the probabilities that the trajectory will be extended at a given step to free lattice nodes.

Introduction of the stated weighting factors does not affect the results obtained for the "thin" filament within the statistical error limits, and in this case, the probability of knot formation can be calculated simply as the ratio of the knotted closed trajectories to the total number of them, as has been done in compiling the table. For a "thick" chain, the weighting factors are very substantial, and they were taken into account in calculating the probabilities of knot formation.

[1)] This can be proved by reducing one of the contour integrals by using Stokes' theorem to an integral over a surface stretched over the contour (see, e.g., [2]). In spite of its great elegance, the Gauss invariant (1) has a number of defects. Thus, there are linkings for which it vanishes (see [1]).

[2)] In the mathematical literature, any closed contour is called a knot. A contour that is topologically equivalent to a circle is called a trivial knot.

[3)] The given relationships hold for all $k=1,\ldots,n$ under the condition of the substitution $n+1\to 1$. Moreover, we assume that the projection of the knot has more than one intersection ($n>1$).

[4)] Certain knots encountered in daily life also have special names. Thus, the know 3_1 is called the trefoil or overhand knot, and the knot 4_1 is called the figure-eight.

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220