

## Topological interactions in systems of mutually interlinked polymer rings

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2004 J. Phys. A: Math. Gen. 37 2881

(<http://iopscience.iop.org/0305-4470/37/8/003>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.66

The article was downloaded on 02/06/2010 at 19:59

Please note that [terms and conditions apply](#).

# Topological interactions in systems of mutually interlinked polymer rings

**Matthias Otto**

Institut für Theoretische Physik, Universität Göttingen, Tammannstrasse 1, D-37077 Göttingen, Germany

E-mail: [otto@theorie.physik.uni-goettingen.de](mailto:otto@theorie.physik.uni-goettingen.de)

Received 8 September 2003

Published 11 February 2004

Online at [stacks.iop.org/JPhysA/37/2881](http://stacks.iop.org/JPhysA/37/2881) (DOI: 10.1088/0305-4470/37/8/003)

## Abstract

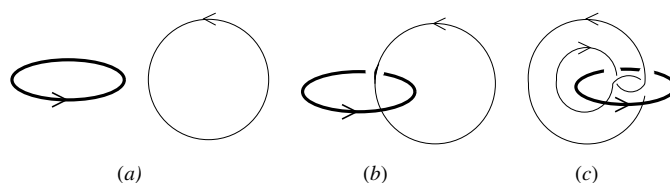
Topological interaction that arises in interlinked polymeric rings such as DNA catenanes is considered. More specifically, the free energy for a pair of linked random walk rings is derived where the distance  $R$  between two segments, each of which is part of a different ring, is kept constant. Topology conservation is imposed by the Gauss invariant. A previous approach (Otto and Vilgis 1998 *Phys. Rev. Lett.* **80** 881) to the problem is refined in several ways. It is confirmed that asymptotically, i.e. for large  $R \gg R_G$  where  $R_G$  is the average size of a single random walk ring, the effective topological interaction (free energy) scales  $\propto R^4$ .

PACS numbers: 02.40.-k, 02.50.-r, 05.20.-y, 05.40.Fb, 82.37.Rs, 87.15.La

## 1. Introduction

Despite their long history, topologically constrained systems such as knots and links became an increasingly popular subject of research in statistical physics [1–6]. Chemically synthesized links (called catenanes) were considered very early [7, 8]. In nature, multi-linked rings made of DNA occur in bacteria such as *Escherichia coli* during the replication process as intermediate products [9]. Specific topological states (links) can be formed artificially by turning enzymes on and off, the so-called topoisomerases, which cut and glue the polymeric rings [9] (see also [10]). These methods have been used to study experimentally the conformational statistics of specific DNA catenanes [11], whose elasticity could also be studied using single-molecule techniques [12]. From the viewpoint of polymer physics topological interactions due to fixed entanglements remain a challenging problem for systems such as rubber networks [13, 14].

A fundamental problem in theoretical approaches to linked polymer rings systems is the specification of topological constraints. The simplest approach introduced by Edwards [1] is a two-ring invariant, known as the Gauss integral. It is one of the simplest invariants



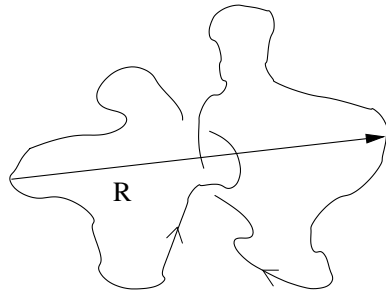
**Figure 1.** (a) The trivial link: two non-entangled rings with linking number 0. (b) Two entangled rings with linking number  $-1$ . (c) Whitehead link: two entangled rings with linking number 0. The oriented surface bounded by the ‘thick’ ring is pierced by the other ring once in a positive sense, once in a negative sense.

involving a double line integral of a function based on respective polymer segment positions of each ring. Thus, it allows us to couple explicitly polymer conformation and topology conservation. Although being a topological invariant, i.e. being independent of the specific polymer conformation as long as the topological state (here the mutual linking number) is conserved, it has the disadvantage of not being one-to-one. The so-called Whitehead link (being entangled due to self-intersections one single ring with itself, see figure 1) and the trivial link (consisting of two non-entangled rings) have both the same linking number zero (see [15] for lucid discussion of this point). This result has aroused criticism against the use of the Gauss invariant in the context of entangled polymer rings (see e.g. [16]).

The present work responds to these objections in the following way. First, we restrict our discussion to random walk rings which are particularly simple to deal with within an analytical approach. In this case, a Whitehead link can be disentangled (justifying a linking number zero), because only the self-entanglement of one ring with itself (see figure 1) keeps the link from falling apart. The second justification for using the Gauss integral is the fact that the Gauss integral appears as the first relevant two-ring invariant appearing within a topological perturbation series constructed from averages (vacuum-to-vacuum expectation values,  $\text{vev}$ 's) of Wilson loops with respect to the non-Abelian Chern–Simons theory [17]. The latter has been shown to be equivalent [18] to the HOMFLY knot polynomial which is an algebraic invariant usually defined in terms of skein relations, i.e. a recursive equation relating different sets of crossings (of the 2D projection of a knot or link) giving a recursive algorithm for transforming different knots and links into each other [17]. In conclusion, the Gauss integral appears as a quantity to define a minimal model for topology conservation which is well defined within the more complete framework just mentioned [19].

More specifically, the present work revisits the problem of calculating the topological interaction for a pair of mutually interlinked polymer rings. This is accomplished by introducing the constraint that the distance between two segments, each belonging to a different ring, be equal to  $\mathbf{R}$  [20]. Previous work [20] on the problem contained several approximations: the distance constraint was not strictly implemented within conformational averages but the distance vector was somehow extracted by a pre-averaging approximation. This approximation can be given up, as the present work will show. Moreover the discrete nature of the linking number is treated within a systematic approach due to Iwata and Kimura [25] (it has recently received some attention in [26]). Perhaps not too surprisingly, the anharmonic attraction  $\propto R^4$  found in [20] still holds for large distance  $R/R_G \gg 1$  where  $R_G \sim \sqrt{NI^2}$  is the radius of gyration of a single random walk ring.

The outline of the paper is as follows. In section 2, we discuss topology conservation using the Gauss invariant together with a distance constraint and define conformational averages. In section 3, the (effective) topological interaction between chain segments belonging to different



**Figure 2.** Two entangled rings with two segments located on different rings kept at a distance  $R$ .

rings is calculated: in section 3.1, we review the topological moment expansion due to Iwata and Kimura [25], in section 3.2 we derive the topological interaction within the approximation of using the second topological moment conditional on a distance constraint  $M_2(R)$ , before finally giving an explicit expression for the latter in section 3.3. Finally we give results in section 3.4 for the topological interaction and discuss its dependence on segment length  $N$ . In section 4, a conclusion and a brief outlook is given.

## 2. Topology conservation with the Gauss invariant and a distance constraint

We consider a very simple system, i.e. a pair of flexible rings which are mutually interlinked as in figure 2 and which are subject to an additional distance constraint. The latter controls the distance between two segments each of which is part of a different ring and is introduced to monitor the effective topological interaction which arises due to the topological constraint.

Based on justifications mentioned in the introduction, the topological constraint imposed on the pair of rings—the fact that they are wound around each other as in figure 2—is expressed in terms of the Gauss invariant

$$\Phi(C_1, C_2) = \Phi_{12} = \frac{1}{4\pi} \oint_{C_1} \oint_{C_2} d\mathbf{r}^1 \wedge d\mathbf{r}^2 \frac{\mathbf{r}^1 - \mathbf{r}^2}{|\mathbf{r}^1 - \mathbf{r}^2|^3}. \quad (1)$$

The vector functions  $\mathbf{r}^\alpha = \mathbf{r}^\alpha(s)$  for  $\alpha = 1, 2$  denote the position in three-dimensional space of segments along the polymer contour, parametrized in terms of  $s$ , for each ring  $C_\alpha$ . The Gauss invariant is always an integer for a given topological state regardless of the deformations imposed on the polymer conformation, i.e. the segment position  $\mathbf{r}^\alpha(s)$ , which leave the topological state invariant (siehe z.B. [17]). The integer which is denoted by  $n$  here is usually called the linking number and counts how often one chain is wound around the other before being closed to a ring (apart from a sign which depends on the respective orientation of each ring).

The problem that we propose to study is the partition sum of a given topological state, i.e. a linking number  $n$ , for a pair of rings subject to the distance constraint as shown in figure 2,

$$Z(n; \mathbf{R}) = \langle \delta(n, \Phi_{12}) \rangle_{\mathbf{R}}. \quad (2)$$

Inside the average to be defined below a Kronecker delta appears which is equal to 1, if the arguments are equal, and 0 else. The average is taken w.r.t. to random walk ring conformations

$$\langle \cdots \rangle_{\mathbf{R}} = \int \mathcal{D}\mathbf{r}^1(s) \int \mathcal{D}\mathbf{r}^2(s) \delta(\mathbf{r}^1(0) - \mathbf{r}^1(N)) \delta(\mathbf{r}^2(0) - \mathbf{r}^2(N)) \\ \times \delta(\mathbf{r}^1(0) - \mathbf{r}^2(0) - \mathbf{R}) \cdots \exp[-(\beta H^1 + \beta H^2)] \frac{1}{Z_{12, \mathbf{R}}} \quad (3)$$

where

$$\beta H^\alpha = \frac{3}{2l^2} \int_0^N ds \left( \frac{\partial \mathbf{r}^\alpha}{\partial s} \right)^2 \quad (4)$$

and  $Z_{12, \mathbf{R}}$  is equal to the numerator on the rhs of equation (3) without the dots. Let us note here that actual averages (carried out in appendix A) involve a discretized version of the continuous formalism given above.

Problem (2) has been studied before by Iwata and Kimura [25] and more recently by Otto and Vilgis [20]. In [25], the integrand of the Gauss invariant was however approximated by its behaviour close to  $|\mathbf{r}^1 - \mathbf{r}^2| \simeq 0$ , an approximation which is not necessary as will be shown below. In [20], the Gauss invariant was treated in full generality, but a full implementation of the distance constraint was circumvented by a pre-averaging procedure involving the tangent vector densities. This approximation can be avoided as will be shown below. The previously obtained results [20] are in fact validated by the subsequent analysis.

The choice of the random walk ensemble for polymer conformations is motivated by the search for an analytical solution. Inclusion of the excluded volume interactions appears possible in principle (in the context of  $O(N)$  field theories [21]), but seems to render the problem of finding an expression for  $Z(n; \mathbf{R})$  rather hopeless. A derivation of topological interactions from the Gauss invariant treating the latter in its most possible generality has not been done up to now even for the simple case of random walk conformations (except for a first approach in [20]).

### 3. The effective topological interaction

#### 3.1. Topological moment expansion

We next proceed to calculate  $Z(n; \mathbf{R})$ . We rewrite the Kronecker delta inside the brackets in equation (2) by introducing a topological charge,

$$Z(n; \mathbf{R}) = \int_{-\pi}^{\pi} \frac{dg}{2\pi} e^{ign} \langle e^{ig\Phi_{12}} \rangle_{\mathbf{R}} \equiv \int_{-\pi}^{\pi} \frac{dg}{2\pi} e^{ign} \tilde{Z}(g; \mathbf{R}). \quad (5)$$

As already noted in [25], the function  $\tilde{Z}(g; \mathbf{R})$  can be expanded in terms of topological moments of order  $k$ ,  $\langle (\Phi_{12})^k \rangle_{\mathbf{R}}$ . Assuming that  $Z(n; \mathbf{R})$  is an even function of  $n$ , only the even moments  $k = 2p$  for an integer  $p$  do contribute. This yields

$$\tilde{Z}(g; \mathbf{R}) = \sum_{p=0}^{\infty} \frac{(ig)^{2p}}{(2p)!} M_{2p}(\mathbf{R}) \quad (6)$$

where

$$M_{2p}(\mathbf{R}) = \langle (\Phi_{12})^{2p} \rangle_{\mathbf{R}}. \quad (7)$$

Next we assume that the  $2p$ -correlation can be decomposed into the products of  $p$  factors involving the correlation  $\langle (\Phi_{12})^2 \rangle_{\mathbf{R}}$  so that

$$\langle (\Phi_{12})^{2p} \rangle_{\mathbf{R}} \simeq \frac{(2p)!}{2^p p!} \langle (\Phi_{12})^2 \rangle_{\mathbf{R}}^p \quad (8)$$

Then one obtains

$$\tilde{Z}(g; \mathbf{R}) = \exp\left(-\frac{g^2}{2}M_2(\mathbf{R})\right). \tag{9}$$

We are thus left with determining the second topological moment given by the distance constraint  $\mathbf{R}$ ,  $M_2(\mathbf{R})$ . Before giving the explicit functional dependence on  $\mathbf{R}$ , we first derive the topological interaction for a given linking number and distance constraint in order to show how  $M_2(\mathbf{R})$  appears in the interaction.

### 3.2. Topological interaction from the second topological moment

In order to establish the effective topological interactions between segments separated by a distance  $R = |\mathbf{R}|$  on different rings which are mutually entangled, we return to the evaluation of the constrained partition sum  $Z(n; \mathbf{R})$  introduced above. The second topological moment  $M_2(R)$  appears—in the approximation (8) given above—inside the conjugate partition sum  $\tilde{Z}(g; \mathbf{R})$ . It now appears natural to directly perform the integral in equation (5). However this leads to negative values for  $Z(n; \mathbf{R})$  for even values of  $n$ , while odd values of  $n$  give positive values, a problem already discussed by Iwata and Kimuara [25]. It is traced back there to an unbalanced distribution of errors involved in the approximation of breaking up higher moments of  $\Phi_{12}^2$  as done in equation (8). Positive errors are accumulated for odd  $n$  and negative errors for even  $n$  [25].

We therefore proceed like in [25], using the ‘continuous Fourier transformation (CFT) method’, whose basic steps are briefly recalled here. While  $Z(n; \mathbf{R})$  is defined for discrete  $n$ , a continuous function  $\zeta(t; \mathbf{R})$  can be obtained by defining

$$\zeta(t; \mathbf{R}) = \sum_n \delta(t - n)Z(n; \mathbf{R}). \tag{10}$$

Its Fourier transform reads as

$$\tilde{\zeta}(u; \mathbf{R}) = \int_{-\infty}^{\infty} dt e^{-iut} \zeta(t; \mathbf{R}). \tag{11}$$

On the level of Fourier transforms,  $\tilde{\zeta}(u; \mathbf{R})$  and  $\tilde{Z}(g; \mathbf{R})$  are identified for  $u = g$  which yields

$$\tilde{\zeta}(u; \mathbf{R}) = \tilde{Z}(u; \mathbf{R}) = \exp\left(-\frac{u^2}{2}M_2(\mathbf{R})\right). \tag{12}$$

The inverse Fourier transform yields

$$\begin{aligned} \zeta(t; \mathbf{R}) &= \int_{-\infty}^{\infty} \frac{du}{2\pi} e^{iut} \tilde{\zeta}(u; \mathbf{R}) \\ &= \frac{1}{\sqrt{2\pi M_2(\mathbf{R})}} \exp\left(-\frac{t^2}{2M_2(\mathbf{R})}\right). \end{aligned} \tag{13}$$

Now, the constrained partition sum of discrete linking numbers  $n \neq 0$  is obtained by integrating

$$Z(n; \mathbf{R}) = \int_{n-\frac{1}{2}}^{n+\frac{1}{2}} dt \zeta(t; \mathbf{R}). \tag{14}$$

In the present case, one finds

$$Z(n; \mathbf{R}) = \frac{1}{2} \left( \operatorname{erf} \left[ \frac{|n| + \frac{1}{2}}{\sqrt{2M_2(\mathbf{R})}} \right] - \operatorname{erf} \left[ \frac{|n| - \frac{1}{2}}{\sqrt{2M_2(\mathbf{R})}} \right] \right) \tag{15}$$

where  $\operatorname{erf}(x)$  is the error function. As noted in [25], the CFT method leads to the following problem. Even though  $\zeta(t, \mathbf{R})$  is assumed to be a multipeak function in  $t$ , the final result just

obtained is a smooth function in  $t$ . In other words the fine structure in  $t$  is lost, which is related to the approximations imposed on  $M_{2p}(R)$  for large  $p \gg 1$  [25]. In the following we are not interested in the exact quantitative dependence of  $Z(n; \mathbf{R})$  on  $n$ , but rather on the asymptotic behaviour for large  $R = |\mathbf{R}|$ . Moreover, the errors contained in the CFT method are evenly distributed among different  $n$ .

The effective topological interaction can be derived from  $Z(n; \mathbf{R})$  as follows:

$$\beta F(n; \mathbf{R}) = -\ln Z(n; \mathbf{R}) \quad (16)$$

where  $\beta = 1/k_B T$  is the inverse thermal energy. If  $M_2(\mathbf{R}) = M_2(R)$  is small and/or  $n \gg 1$  which happens for large  $R = |\mathbf{R}|$  (see below), such that  $x_{\pm} = \frac{|n| \pm \frac{1}{2}}{\sqrt{2M_2(\mathbf{R})}} \gg 1$  then the following expansion will be used:

$$\beta F(n; \mathbf{R}) = \beta F(n; R) = \frac{(|n| - \frac{1}{2})^2}{2M_2(R)} + \ln \left[ \frac{|n| - \frac{1}{2}}{\sqrt{2\pi M_2(R)}} \right] + \mathcal{O}(1/x_-). \quad (17)$$

We see that the behaviour of the topological interaction  $\beta F(n; R)$  w.r.t.  $R$ , in particular for  $R \gg l\sqrt{N}$ , is dominated by  $1/M_2(R)$ .

### 3.3. The second topological moment

We now proceed to calculate  $M_2(\mathbf{R})$ . Using the Fourier integral representation of the Gauss invariant

$$\Phi_{12} = -i \int_{\mathbf{q}} \int_0^N ds \int_0^N ds' \dot{r}_{\mu}^1(s) \dot{r}_{\nu}^2(s') \exp(i\mathbf{q} \cdot (\mathbf{r}^1(s) - \mathbf{r}^2(s'))) \epsilon_{\mu\nu\lambda} \frac{q_{\lambda}}{q^2} \quad (18)$$

where summation over repeated indices is implied and  $\int_{\mathbf{q}} \dots = \int d^3q / (2\pi)^3 \dots$ , the second topological moment reads as

$$M_2(\mathbf{R}) = - \int_{\mathbf{q}} \int_{\mathbf{k}} \Pi_{\alpha=1}^4 \left( \int_0^N ds_{\alpha} \right) \epsilon_{\mu\nu\lambda} \epsilon_{\sigma\tau\rho} \frac{q_{\lambda} k_{\rho}}{q^2 k^2} \langle \dot{r}_{\mu}^1(s_1) \dot{r}_{\nu}^2(s_3) \dot{r}_{\sigma}^1(s_2) \dot{r}_{\tau}^2(s_4) \times \exp(i\mathbf{q} \cdot (\mathbf{r}^1(s_1) - \mathbf{r}^2(s_3))) \exp(i\mathbf{k} \cdot (\mathbf{r}^1(s_2) - \mathbf{r}^2(s_4))) \rangle_{\mathbf{R}}. \quad (19)$$

The term in brackets inside the integrals can be rewritten as

$$\gamma_{\mu\nu\sigma\tau} = \langle \dot{r}_{\mu}^1(s_1) \dot{r}_{\nu}^2(s_3) \dot{r}_{\sigma}^1(s_2) \dot{r}_{\tau}^2(s_4) \exp(i\mathbf{q} \cdot (\mathbf{r}^1(s_1) - \mathbf{r}^2(s_3))) \exp(i\mathbf{k} \cdot (\mathbf{r}^1(s_2) - \mathbf{r}^2(s_4))) \rangle_{\mathbf{R}} \\ = \exp(i(\mathbf{q} + \mathbf{k}) \cdot \mathbf{R}) F_{\mu\sigma}(\mathbf{q}, \mathbf{k}, s_1, s_2) F_{\nu\tau}(-\mathbf{q}, -\mathbf{k}, s_3, s_4). \quad (20)$$

The factorization on the rhs is due to the factorization of conformational averages with respect to polymers 1 and 2 in equation (3). Details are given in appendix A. We find for  $F_{\mu\sigma}(\mathbf{q}, \mathbf{k}, s_1, s_2)$  that

$$F_{\mu\sigma}(\mathbf{q}, \mathbf{k}, s_1, s_2) = \frac{1}{2} \left\{ \frac{l^2}{3} \delta_{\mu\sigma} \left( \frac{1}{N} - \delta(s_1 - s_2) \right) + \left( \frac{l^2}{3} \right)^2 \left[ \frac{1}{N} (s_1 q_{\sigma} + s_2 k_{\sigma}) - k_{\sigma} \right] \right. \\ \left. \times \left[ \frac{1}{N} (s_1 q_{\mu} + s_2 k_{\mu}) - q_{\mu} - k_{\mu} \right] \right\} \\ \times \exp \left\{ -\frac{l^2}{6N} \left[ s_1 (\mathbf{q} + \mathbf{k})^2 + (s_2 - s_1) k^2 - \frac{1}{N} (s_1 \mathbf{q} + s_2 \mathbf{k})^2 \right] \right\} \\ + [s_1 \leftrightarrow s_2, \mathbf{q} \leftrightarrow \mathbf{k}]. \quad (21)$$

The last line on the rhs indicates the operations to be done in order to obtain a fully symmetrized expression (see appendix A). Let us note first that the term following the factor  $\left(\frac{l^2}{3}\right)^2$  on the

rhs does not contribute to the second moment, because expressions such as  $\dots q_\mu q_\lambda \epsilon_{\mu\nu\lambda} \dots$  vanish. In order to further simplify the subsequent analysis, we neglect terms involving  $1/N$  as we assume  $N \gg 1$  as in [23]. Then  $F_{\mu\sigma}(\mathbf{q}, \mathbf{k}, s_1, s_2)$  reads as

$$F_{\mu\sigma}(\mathbf{q}, \mathbf{k}, s_1, s_2) \simeq -\left(\frac{l^2}{3}\right) \delta_{\mu\sigma} \delta(s_1 - s_2) \exp\left\{-\frac{l^2}{6} s_1 (\mathbf{q} + \mathbf{k})^2\right\}. \quad (22)$$

The second topological moment may then be computed as

$$M_2(\mathbf{R}) = -2 \left(\frac{l^2}{3}\right)^2 \int_{\mathbf{q}} \int_{\mathbf{k}} f(\mathbf{q} + \mathbf{k}, N)^2 \frac{\mathbf{q} \cdot \mathbf{k}}{q^2 k^2} e^{i(\mathbf{q} + \mathbf{k}) \cdot \mathbf{R}} \quad (23)$$

where

$$f(\mathbf{u}, N) = \int_0^N ds e^{-\frac{l^2}{6} s \mathbf{u}^2} = \frac{1}{\left(\frac{l^2}{3}\right) \mathbf{u}^2} (1 - e^{-\frac{l^2 N}{6} \mathbf{u}^2}). \quad (24)$$

Transforming  $\mathbf{k} \rightarrow \mathbf{u} = \mathbf{q} + \mathbf{k}$ , one obtains

$$M_2(\mathbf{R}) = -8 \int_{\mathbf{q}} \int_{\mathbf{u}} \frac{\mathbf{q} \cdot (\mathbf{u} - \mathbf{q})}{q^2 |\mathbf{u} - \mathbf{q}|^2} \frac{e^{i\mathbf{u} \cdot \mathbf{R}}}{u^4} (1 - e^{-\frac{l^2 N}{6} u^2})^2. \quad (25)$$

Introducing  $\mathbf{u}' = \sqrt{l^2 N / (6)} \mathbf{u}$  and  $\mathbf{X} = \mathbf{R} / \sqrt{l^2 N / (6)}$  and dropping primes, the double integral is slightly simplified:

$$M_2(\mathbf{R}) = -8 \int_{\mathbf{q}} \int_{\mathbf{u}} \frac{\mathbf{q} \cdot (\mathbf{u} - \mathbf{q})}{q^2 |\mathbf{u} - \mathbf{q}|^2} \frac{e^{i\mathbf{u} \cdot \mathbf{X}}}{u^4} (1 - e^{-u^2})^2. \quad (26)$$

The integration which involves some approximations affecting the short-distance behaviour (to be discussed below) and which is given in appendix B, yields

$$M_2(R) = \frac{1}{8\pi^3} \left( \Lambda \frac{\sqrt{\pi}}{2} - \pi {}_1F_1\left(-\frac{1}{2}; \frac{3}{2}; \frac{X^2}{4}\right) \right) e^{-X^2/4} \quad (27)$$

where  $R = X \sqrt{l^2 N / 6}$  and  ${}_1F_1(\alpha; \beta; x)$  is the confluent hypergeometric function. The expression involves a cutoff  $\Lambda \sim \sqrt{N}$  (see appendix A) whose relevance will be discussed below. It is interesting to note what happens when  $M_2(R)$  is integrated over all distances between the segments. Then the unconditional second topological moment is obtained as

$$\begin{aligned} M_2 &= V^{-1} \int d^3 R M_2(R) \\ &= V^{-1} \sqrt{l^2 N / 6}^3 4\pi \int_0^\infty dX X^2 \frac{1}{8\pi^3} \left( \Lambda \frac{\sqrt{\pi}}{2} - \pi {}_1F_1\left(-\frac{1}{2}; \frac{3}{2}; \frac{X^2}{4}\right) \right) e^{-X^2/4} \\ &\sim V^{-1} l^3 N^{3/2} (c_1 N^{1/2} - c_2) \end{aligned} \quad (28)$$

where in the last line  $\Lambda \sim \sqrt{N}$  has been used and where  $c_1$  and  $c_2$  are numerical factors. Note that the factor  $1/V$  on the rhs of the previous equation is understood in the limits  $V \rightarrow \infty$  and  $N \rightarrow \infty$  such that  $\rho = N/V$  is the density. This situation is given if two rings fill out a macroscopic volume. Then the unconditional second topological moment has the scaling

$$M_2 \sim \rho l^3 (c_1 N - c_2 N^{1/2}) \quad (29)$$

a result which up to numerical factors  $c_1$  and  $c_2$  has been found for the topological moment of randomly entangled polymer rings in a dense system where the excluded volume effect is ignored [22, 24]. Therefore, the cutoff-dependent first term on the rhs of equation (27) appears essential in order to establish consistency with the well-known result for  $M_2$  for random walk rings to scale to dominant order like  $\propto N$ . We will see below that it affects the effective segment–segment interaction only for small, up to intermediate, deformations of the rings.



As shown in the previous section, the behaviour of  $1/M_2(R)$  is particularly important to obtain the large- $R$  behaviour of the topological interaction. Note that

$${}_1F_1\left(-\frac{1}{2}; \frac{3}{2}; \frac{X^2}{4}\right) = e^{X^2/4} {}_1F_1\left(2; \frac{3}{2}; -\frac{X^2}{4}\right) \quad (30)$$

and using for large  $X \gg 1$

$${}_1F_1\left(2; \frac{3}{2}; -\frac{X^2}{4}\right) \simeq -\frac{4}{X^4} \left(1 + \frac{12}{X^2} + \dots\right) \quad (31)$$

one obtains:

$$\begin{aligned} \frac{1}{M_2(R)} &\simeq 8\pi^3 \left(\frac{\sqrt{\pi}}{2} \Lambda \exp(-X^2/4) + \frac{4\pi}{X^4}\right)^{-1} \\ &\simeq 2\pi^2 X^4 \end{aligned} \quad (32)$$

for large  $X = R/\sqrt{Nl^2/6}$ . It gives the following effective topological interaction:

$$\beta F(n; X) \simeq \pi^2 \left(n - \frac{1}{2}\right)^2 X^4. \quad (33)$$

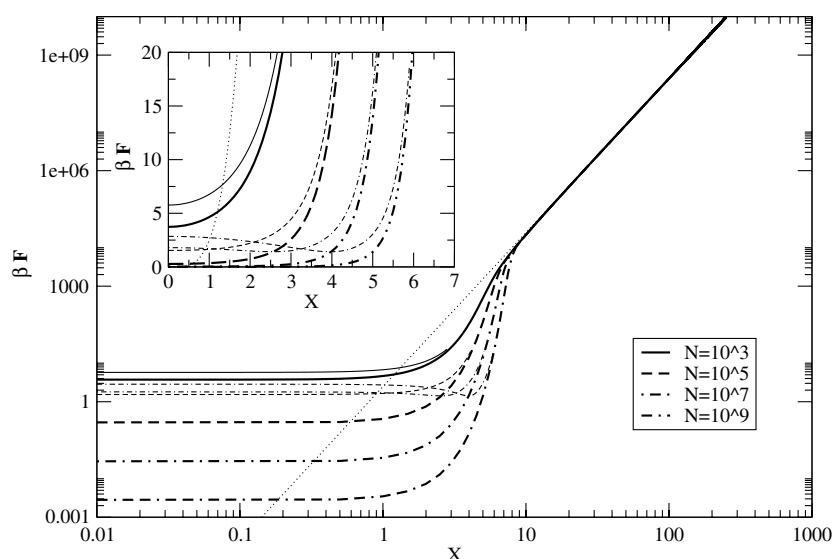
Concerning the  $X^4$  dependence, this result agrees perfectly with the previously derived asymptotics for the topological interaction of two concatenated chains based on a pre-averaging approximation [20].

### 3.4. Results

Given the expression for the conditional topological moment (27), we now consider the effective topological interaction in more detail. For large  $X$ , we use the first term in the expansion (17). For small  $X$  where equation (17) is bound to fail, the exact expression (15) for the partition function (based on the second topological moment conditional on  $R$  or  $X$ ) is used. In figure 3, curves for small  $X$  (obtained numerically from equation (15)) for the topological interaction  $\beta F$  for linking number  $n = 1$  for various chain lengths  $N$  are shown, together with the corresponding curves for all  $X$  calculated from the first term in equation (17), with both sets of curves involving equation (27) for the second topological moment  $M_2(R)$ .

The topological interactions computed from the full expression (15) smoothly approach the approximate ones (valid for large  $X$ ) based on equation (17). Whereas for large  $X$  the topological interaction is independent of the chain length  $N$  approaching the asymptotic form (33), such a dependence appears for intermediate and small  $X$ . To be precise, the interaction becomes weaker for larger  $N$  for a given intermediate  $X$ , just below  $X = 10$ . This effect might be due to the discrete polymer model used here. Let us consider two systems of two concatenated rings which are similar in their conformation but which have different chain lengths  $N_1$  and  $N_2$  with  $N_2 > N_1$ . Then the system with chain length  $N_2$  suffers a smaller entropy loss due to the topological constraint. Thus the topological interaction is reduced.

However, for very small  $X$  the topological interaction does not show a monotonous dependence on  $N$ , but passes through a minimum as  $N$  is increased from  $10^3$  to  $10^9$ . Moreover, for very large  $N$  a minimum of the topological interaction as a function of  $X$  starts to develop. A strong repulsion of segments belonging to different rings at small  $X$  found in [20] is not confirmed by the present calculation. On the other hand, the present work depends on approximations (see appendix B) which possibly influence the short-distance behaviour of the topological interaction. Therefore, no conclusive statement is claimed for the topological interactions derived from the Gauss invariant on small scales  $X$ . A more complete analysis of the short-distance behaviour is necessary and is the subject of future work.



**Figure 3.** The topological interactions for linking number  $n = 1$  based on, one, the first term in equation (17) (thick lines), and two, on the full expression (15) for the partition function (thin lines), both together with equation (27). The thin dotted line corresponds to the asymptotic expression (33). The inset shows in a double-linear plot how the curves based on the full expression (15) approach the approximate ones (equation (17)).

For extreme elongations  $X \gg 1$ , one might expect for the topological interaction a crossover from  $X^4$  to  $X^2$  as claimed in [26], based on the argument that when two random walk rings linked with linking number 1 are fully stretched, they form an effective random walk chain. Apparently, this limit cannot be reached by the present approach. The argument just given seems to assume, however, some local gluing and some partial inextensibility of the chains which then leads to a mutual attachment of the rings due to the topological constraint. Each ring conformation is however treated here w.r.t. to the ensemble of random walks. Therefore the effective conformation of a single random walk chain is not reached.

#### 4. Discussion

In the present work, the topological interaction between segments belonging to different polymer rings which are mutually interlinked is considered. Two segments are singled out and kept at a distance  $R$ , while the conformations of each ring are averaged w.r.t. a random walk ensemble. Supplementing a previous calculation [20], the present analysis introduces several refinements. First, the distance constraint is implemented from the very beginning. Second, the discrete nature of the linking number is treated systematically following [25].

The topological interaction behaves asymptotically, i.e. for  $X \gg 1$  where  $X = R/\sqrt{Nl^2}/6$ , as  $\sim X^4$  as found previously [20]. For smaller  $X \leq 10$ , the topological interaction depends on the chain length  $N$ . For intermediate distances  $X$ , an argument related to the reduced entropy loss due to the topology conservation for increasing  $N$  can be given. For small  $X$ , no final conclusive statement on the topological interaction can be given, as the approximations used for the conformational averages in order to obtain analytical results possibly affect small length scales. We remark however that the present result is consistent with calculations of the unconditional second topological moment [22, 24].

The present work may be considered as a further step towards understanding the elasticity of DNA catenanes (interlinked rings) which can be studied in principle using single-molecule techniques [12]. A first step was the previous study [20] in order to understand the conformational statistics of DNA catenanes [11]. The simple random walk model used here is of course not realistic in this context. Apart from the excluded volume problem, DNA molecules are semiflexible chains [27] which require the Kratky–Porod model [28] that however leads to analytical problems. Nonetheless, for long chains, self-avoiding walk behaviour is recovered.

Possible directions of research on the problem presented here do not only concern the use of more realistic conformational models relevant to the study of biomolecules. Future work should aim at a more accurate treatment of conformational averages, in particular the inclusion of terms which were omitted here for large  $N$  (which amount to neglecting the closure of chains). Another important problem is the issue of localization: i.e. the hypothesis, that a number of segments are localized close to a region containing the link, motivating concepts such as slip-links (see e.g. [29]) which have found renewed interest [30].

### Acknowledgment

Financial support by the DFG under grant Zi209/6-1 is gratefully acknowledged.

### Appendix A. The correlation function $\gamma_{\mu\nu\sigma\tau}$

Following [23], we calculate the following correlation function of equation (20):

$$\gamma_{\mu\nu\sigma\tau} = \left\langle \dot{r}_\mu^1(s_1) \dot{r}_\nu^2(s_3) \dot{r}_\sigma^1(s_2) \dot{r}_\tau^2(s_4) \exp(i\mathbf{q} \cdot (\mathbf{r}^1(s_1) - \mathbf{r}^2(s_3))) \exp(i\mathbf{k} \cdot (\mathbf{r}^1(s_2) - \mathbf{r}^2(s_4))) \right\rangle_{\mathbf{R}}. \quad (\text{A.1})$$

To do so, we introduce a discretized random walk  $\mathbf{r}^1(s) = \mathbf{R}_0^1 + \sum_{i=1}^s \mathbf{b}_i^1$  such that the measure of the path integral in equation (3) becomes

$$\mathcal{D}\mathbf{r}^1(s) \rightarrow \int d^3 R_0^1 \prod_{i=1}^N \int d^3 b_i^1. \quad (\text{A.2})$$

Then the correlation function given above is derived from the following generating functional:

$$G(\{\mathbf{h}_i^1\}, \{\mathbf{h}_i^2\}; \mathbf{q}, \mathbf{k}; s_1, s_2, s_3, s_4) = \langle \exp(i\mathbf{q} \cdot (\mathbf{r}^1(s_1) - \mathbf{r}^2(s_3))) \exp(i\mathbf{k} \cdot (\mathbf{r}^1(s_2) - \mathbf{r}^2(s_4))) \rangle_{\{\mathbf{R}\}, \{\mathbf{h}_i^1\}, \{\mathbf{h}_i^2\}}. \quad (\text{A.3})$$

The average is understood as

$$\begin{aligned} G(\{\mathbf{h}_i^1\}, \{\mathbf{h}_i^2\}; \mathbf{q}, \mathbf{k}; s_1, s_2, s_3, s_4) &= \frac{1}{Z_{12, \mathbf{R}}} \int d^3 R_0^1 \prod_i \int d^3 b_i^1 \int d^3 R_0^2 \prod_i \int d^3 b_i^2 \\ &\times \delta\left(\sum_i \mathbf{b}_i^1\right) \delta\left(\sum_i \mathbf{b}_i^2\right) \delta(\mathbf{R}_0^1 - \mathbf{R}_0^2 - \mathbf{R}) \\ &\times \exp\left[-\beta H^1 - \beta H^2 + \sum_{\alpha=1,2} \sum_i \mathbf{h}_i^\alpha \cdot \mathbf{b}_i^\alpha\right] \exp[-i(\mathbf{q} + \mathbf{k}) \cdot \mathbf{R}_0^1 + i(\mathbf{q} + \mathbf{k}) \cdot \mathbf{R}_0^2] \\ &\times \exp\left[-i\mathbf{q} \cdot \sum_{i=1}^{s_1} \mathbf{b}_i^1 - i\mathbf{k} \cdot \sum_{i=1}^{s_2} \mathbf{b}_i^1 + i\mathbf{q} \cdot \sum_{i=1}^{s_3} \mathbf{b}_i^2 + i\mathbf{k} \cdot \sum_{i=1}^{s_4} \mathbf{b}_i^2\right]. \quad (\text{A.4}) \end{aligned}$$

Products and sums in the above equation marked by an index  $i$  are only supposed to cover the range  $i = 1, \dots, N$ . The integrations w.r.t. to  $\mathbf{R}_0^\alpha$  for  $\alpha = 1, 2$  in the numerator and denominator on the rhs of the above equation are easily carried out to give

$$G(\{\mathbf{h}_i^1\}, \{\mathbf{h}_i^2\}; \mathbf{q}, \mathbf{k}; s_1, s_2, s_3, s_4) = \exp(i(\mathbf{q} + \mathbf{k}) \cdot \mathbf{R}) F(\{\mathbf{h}_i^1\}, \mathbf{q}, \mathbf{k}, s_1, s_2) \times F(\{\mathbf{h}_i^2\}, -\mathbf{q}, -\mathbf{k}, s_3, s_4) \quad (\text{A.5})$$

where

$$F(\{\mathbf{h}_i\}, \mathbf{q}, \mathbf{k}, s_1, s_2) = \frac{1}{Z_1} \prod_i \int d^3 b_i \delta\left(\sum_i \mathbf{b}_i\right) \exp\left[-\frac{3}{2l^2} \sum_i (\mathbf{b}_i)^2 + \sum_i \mathbf{h}_i \cdot \mathbf{b}_i\right] \times \exp\left[-i\mathbf{q} \cdot \sum_{i=1}^{s_1} \mathbf{b}_i - i\mathbf{k} \cdot \sum_{i=1}^{s_2} \mathbf{b}_i\right]. \quad (\text{A.6})$$

The partition sum  $Z_1$  is the single ring partition function divided by a volume factor (originating from the integrations w.r.t. to  $\mathbf{R}_0^\alpha$  for  $\alpha = 1, 2$  which have already been carried out). Now one has to choose an order for  $s_1, s_2$  which are now considered as being discrete out of  $1, \dots, N$ , say  $s_1 \leq s_2$ . Then the integrations are easily carried out to give

$$F(\{\mathbf{h}_i\}, \mathbf{q}, \mathbf{k}, s_1, s_2) = \exp\left[\frac{l^2}{6N} \left(\sum_i \mathbf{h}_i + s_1 \mathbf{q} + s_2 \mathbf{k}\right)^2 - \frac{l^2}{6} \left(\sum_{i=1}^{s_1} (\mathbf{h}_i + \mathbf{q} + \mathbf{k})^2 + \sum_{i=s_1+1}^{s_2} (\mathbf{h}_i + \mathbf{k})^2 + \sum_{i=s_2+1}^N \mathbf{h}_i^2\right)\right]. \quad (\text{A.7})$$

The case  $s_1 > s_2$  is obtained by interchanging  $s_1 \leftrightarrow s_2$  and  $\mathbf{q} \leftrightarrow \mathbf{k}$ . Now, the correlation function  $\gamma_{\mu\nu\sigma\tau}$  is obtained as follows:

$$\gamma_{\mu\nu\sigma\tau} = \frac{1}{(-i)^4} \frac{\partial}{\partial h_{\mu, s_1}^1} \frac{\partial}{\partial h_{\nu, s_3}^2} \frac{\partial}{\partial h_{\sigma, s_2}^1} \frac{\partial}{\partial h_{\tau, s_4}^2} G(\{\mathbf{h}_i^1\}, \{\mathbf{h}_i^2\}; \mathbf{q}, \mathbf{k}; s_1, s_2, s_3, s_4) \Big|_{\mathbf{h}_i^1=0, \mathbf{h}_i^2=0}. \quad (\text{A.8})$$

Using equation (A.5) together with equation (A.7) one obtains equation (20) with equation (21) for  $\gamma_{\mu\nu\sigma\tau}$  given in the main text.

## Appendix B. Approximate evaluation of integrals

Defining the integral w.r.t.  $\mathbf{q}$  in equation (26) as  $l(\mathbf{u})$  one obtains

$$M_2(\mathbf{R}) = -\frac{16}{(2\pi)^2} \frac{1}{X} \int_0^\infty du \frac{\sin(uX)}{u^3} (1 - e^{-u^2})^2 l(u) \quad (\text{B.1})$$

where

$$l(u) = l(\mathbf{u}) = \int_{\mathbf{q}} \frac{\mathbf{q} \cdot (\mathbf{u} - \mathbf{q})}{q^2 |\mathbf{u} - \mathbf{q}|^2} = \frac{2}{(2\pi)^2} B(3/2, 3/2) \int_0^\Lambda dq \frac{u^2 q^2 - q^4}{(q^2 + u^2)^2}. \quad (\text{B.2})$$

Note that the integral in the last equation needs to be cut off by a parameter  $\Lambda$  which is related to the cutoff in momentum space  $\Lambda'$  via  $\Lambda = \Lambda' \sqrt{l^2 N}/6$  and thus proportional to  $\sqrt{N}$  if  $\Lambda' \sim l^{-1}$ . Evaluating the  $q$ -integral yields for  $l(u)$

$$l(u) = \frac{1}{(2\pi)^2} \frac{\pi}{8} \left( \frac{\pi}{2} u - 2\Lambda + 3u \arctan(\Lambda/u) - \frac{\Lambda u^2}{\Lambda^2 + u^2} \right). \quad (\text{B.3})$$

In order to make analytical progress, let us approximate  $(1 - e^{-u^2})^2/u^3 \simeq u e^{-u^2}$  in equation (26). The error involved in this approximation concerns large  $u$ , and the short-distance behaviour of  $M_2$ , which is discussed in the main text. Then the second topological moment is given by the expression

$$M_2(R) = -\frac{16}{(2\pi)^2} \frac{1}{X} \int_0^\infty du l(u) u \sin(uX) e^{-u^2}. \quad (\text{B.4})$$

Inside the integral  $l(u)$  is approximated, for  $\Lambda \gg 1$ , as

$$l(u) \simeq \frac{1}{(2\pi)^2} \frac{\pi}{8} \left( -2\Lambda + 2\pi u - \frac{u^2}{\Lambda} \right) \quad (\text{B.5})$$

where  $\arctan(\Lambda/u) \simeq \pi/2$  for  $\Lambda \gg 1$ . Using the above approximation, evaluation of the integral w.r.t.  $u$  involves the following integrals. The term of order 0 in  $u$  gives the integral

$$I_1 = \int_0^\infty du u \sin(uX) e^{-u^2} = \frac{\sqrt{\pi} X}{4} e^{-X^2/4}. \quad (\text{B.6})$$

The term linear in  $u$  in  $l(u)$  yields the integral

$$I_2 = \int_0^\infty du u^2 \sin(uX) e^{-u^2} = \frac{X}{2} e^{-X^2/4} {}_1F_1 \left( -\frac{1}{2}; \frac{3}{2}; \frac{X^2}{4} \right) \quad (\text{B.7})$$

while the term of order 2 in  $u$  involves evaluating

$$I_3 = \int_0^\infty du u^3 \sin(uX) e^{-u^2} = \frac{X}{2} e^{-X^2/4} \Gamma(5/2) {}_1F_1 \left( -1; \frac{3}{2}; \frac{X^2}{4} \right). \quad (\text{B.8})$$

The function  ${}_1F_1(-1; \frac{3}{2}; \frac{X^2}{4})$  explicitly gives  $1 - X^2/6$ . However, we will neglect the  $u^2$  term in  $l(u)$  because it is multiplied by a factor  $1/\Lambda$  (see equation (B.5)). Collecting terms, one obtains for the second moment

$$M_2(R) = \frac{1}{8\pi^3} \left( \Lambda \frac{\sqrt{\pi}}{2} - \pi {}_1F_1 \left( -\frac{1}{2}; \frac{3}{2}; \frac{X^2}{4} \right) \right) e^{-X^2/4} \quad (\text{B.9})$$

where  $R = X\sqrt{l^2 N/6}$  and  ${}_1F_1(\alpha; \beta; x)$  is the confluent hypergeometric function (see equation (27) in the main text).

## References

- [1] Edwards S F 1968 *J. Phys. A:Math. Gen.* **1** 15
- [2] de Gennes P G 1979 *Scaling Concepts in Polymer Physics* (Ithaca, NY: Cornell University Press)
- [3] Nechaev S K 1996 *Statistics of Knots and Entangled Random Walks* (Singapore: World Scientific)
- [4] Kholodenko A L and Vilgis T A 1998 *Phys. Rep.* **298** 251
- [5] Grosberg A Y 2000 *Phys. Rev. Lett.* **85** 3585
- [6] Metzler R, Hanke A, Dommersnes P G, Kantor Y and Kardar M 2002 *Phys. Rev. Lett.* **88** 188101  
Farago O, Kantor Y and Kardar M 2002 *Europhys. Lett.* **60** 53
- [7] Wasserman E 1960 *J. Am. Chem. Soc.* **82** 4433
- [8] Frisch H L and Wasserman E 1962 *J. Am. Chem. Soc.* **83** 3789
- [9] Wasserman S A and Cozzarelli N R 1986 *Science* **232** 951
- [10] Alberts B *et al* 2002 *The Molecular Biology of the Cell* (New York: Garland)
- [11] Levene S D, Donahue C, Boles T C and Cozzarelli N R 1995 *Biophys. J.* **69** 1036
- [12] Arai Y, Yasuda R, Akashi K-I, Harada Y, Miyata H, Kinoshita K Jr and Itoh H 1999 *Nature* **339** 446
- [13] Kremer K and Grest G 1995 *Phys. World* (March) 39
- [14] Everaers R and Kremer K 1995 *Macromolecules* **28** 7291  
Everaers R and Kremer K 1995 *Phys. Rev. E* **53** R37  
Everaers R and Kremer K 1996 *Anomalous Diffusion. From Basics to Applications Proc. 11th Max Born Symp.*  
ed A Pekalski and K Sznajd-Weron (Berlin: Springer) 221

- [15] Brereton M G and Shah S 1980 *J. Phys. A: Math. Gen.* **13** 2751
- [16] Nechaev S K and Rostiasvili V G 1993 *J. Phys. II France* **3** 91
- [17] Guadagnini E 1993 *The Link Invariants of the Chern–Simons Field Theory* (Berlin: de Gruyter&Co)
- [18] Witten E 1989 *Commun. Math. Phys.* **121** 351
- [19] Otto M 1996 Die statistische Mechanik flexibler, verschlaufte und unverschlaufte Polymerringe (in German)  
*PhD Thesis* Universität Mainz (available at <http://www.Theorie.Physik.UNI-Goettingen.DE/~otto/pubs.html>)
- [20] Otto M and Vilgis T A 1998 *Phys. Rev. Lett.* **80** 881
- [21] des Cloizeaux J and Jannink G 1990 *Polymers in Solution: Their Modelling and Structure* (Oxford: Clarendon)
- [22] Ferrari F, Kleinert H and Lazzizzera I 2000 *Phys. Lett. A* **276** 31  
Ferrari F, Kleinert H and Lazzizzera I 2000 *Eur. Phys. J. B* **18** 645–54
- [23] Brereton M G and Vilgis T A 1992 *Phys. Rev. A* **45** 7413
- [24] Otto M 2001 *J. Phys. A: Math. Gen.* **34** 2539
- [25] Iwata K and Kimura T 1981 *J. Chem. Phys.* **74** 2039
- [26] Brereton M G 2001 *J. Phys. A: Math. Gen.* **34** 5131
- [27] Marko J F and Siggia E D 1994 *Science* **265** 506  
Marko J F and Siggia E D 1995 *Phys. Rev. E* **52** 2912  
Marko J F and Siggia E D 1995 *Macromolecules* **28** 8759
- [28] Kratky O and Porod G 1949 *Rec. Trav. Chim.* **68** 1106
- [29] Edwards S F and Vilgis T A 1988 *Rep. Prog. Phys.* **51** 243
- [30] Metzler R, Hanke A, Dommersnes P G, Kantor Y and Kardar M 2002 *Phys. Rev. E* **65** 061103