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Topological entanglement in polymers

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Abstract. The Brereton–Shah problem of two polymer loops topologically linked is studied by field theoretical methods in the limit that one polymer is allowed to fill a macroscopic volume at finite density $\bar{\rho}$. For $L \gg 1$ it is shown that typically for winding numbers $m^2 \ll L^\alpha \bar{\rho}$, $\alpha = 2 - \varepsilon\nu$, universality is observed with the critical exponents of the unconstrained system despite the presence of a ‘dangerous’ renormalisation group instability. Here L is the length of the smaller polymer loop whose mean square size $\langle R^2 \rangle \sim L^{2\nu}$; $\nu = \frac{1}{2}$ for random flight statistics or $\nu \sim \frac{3}{5}$ for the swelled chain case.

1. Introduction

Over the years the study of linear polymer chains by means of a direct isomorphism with a variety of $n \rightarrow 0$ field theories, outlined by many authors (de Gennes 1972, des Cloizeaux 1975, Emery 1975, Burch and Moore 1976), has resulted in a reasonably complete understanding of such systems. In particular, these field theoretic approaches present an ideal framework within which to discuss the universality structure of polymer systems using the renormalisation group and the ε expansion. Despite these great advances, the statistical mechanics of polymer loops has in contrast remained relatively undeveloped, for the notion of ‘topological entanglement’ presents many new problems. Edwards (1967, 1968) in a series of papers, and more recently Brereton and Shah (1980, BS), have outlined an approach to the problem of including the natural topological constraints on such systems, and have demonstrated an isomorphism between topologically constrained polymer models and a series of $n \rightarrow 0$ locally gauge-invariant field theories. However, beyond some simple observations by Edwards based on the renormalisability of such models, the universality class structure has not been discussed in the literature. We shall in this paper seek to remedy this shortcoming for the BS system of two topologically linked polymer loops, showing that although the renormalisation group reveals a dangerous instability, the associated bare coupling is asymptotically ($L \gg 1$) very small, so that universality is observed with the critical exponents of the linear polymer system.

Following a brief discussion of the knot classification problem and its role in the physics of polymer loops in § 2, we shall in § 3 outline the origin of the isomorphism which exists between the BS model of two coupled polymer loops and an $n \rightarrow 0$ locally gauge-invariant field theory. Formulated as a field theory, we argue that it is useful to consider a continuation of the entanglement problem to d spatial dimensions. In §§ 4 and 5 we shall discuss the implications of this continuation for the problem of long topologically entangled polymer loops governed by excluded volume statistics, first

using naive perturbation theory and then by means of the more sophisticated renormalisation group approach. Particular emphasis will be placed on the entropy S_m and the mean square polymer size $\langle R^2 \rangle_m$ in the presence of a topological constraint specified by an index m .

2. The statistical mechanics of polymer loops

Topological entanglements represent an invariant aspect of a polymer system for they are by definition conserved throughout all the configurational changes that the polymers undergo. A simple example is provided by two polymer loops, for we see that two loops initially unlinked cannot in the course of time become linked without breaking one of the loops.

A proper statistical description of a system of polymer loops must therefore, in addition to the usual physical variables, include a complete set of topological variables which serve to distinguish between all configurations which are not topologically equivalent. The topological variables are of course quenched in the sense that they are determined solely by the initial topological state of the system. Unfortunately the classification of knots (entanglements) is horrendously complex, so we shall be forced to follow Edwards and opt for a partial classification in terms of the Gauss integral I given by Alexandroff and Hopf (1935):

$$I_{\alpha\beta} = I_{\alpha\beta}(\mathcal{C}_\alpha, \mathcal{C}_\beta) = \frac{1}{4\pi} \iint_{\mathcal{C}_\alpha, \mathcal{C}_\beta} d\mathbf{r}_\alpha \times d\mathbf{r}_\beta \cdot \nabla \left(\frac{1}{|\mathbf{r}_\alpha - \mathbf{r}_\beta|} \right). \tag{2.1}$$

Here $\mathcal{C}_\alpha, \mathcal{C}_\beta$ are two polymer loops parametrised by vectors $\mathbf{r}_\alpha, \mathbf{r}_\beta$ in three dimensions. The function $I_{\alpha\beta}$ taking values among the integers m , positive and negative, is indeed a true topological invariant for its value is unchanged under all deformations for which the loops are unbroken. Examining figure 1, it is clear that $I_{\alpha\beta}$ is a valuable invariant, although it indeed only represents a partial classification:

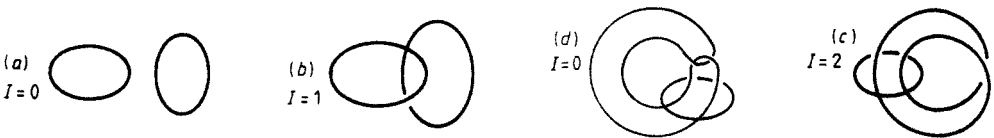


Figure 1.

Later we shall propose a continuation of our problem to d spatial dimensions, so it is important to remark here that of course the physical problem exists only in $d = 3$ since both for $d = 2$ and $d \geq 4$ the problem becomes trivial.

Despite the impracticability of obtaining a full topological specification, some functions of physical importance may be studied in the presence of entanglements. Consider for example a system of two polymer loops $\mathcal{C}_\alpha, \mathcal{C}_\beta$ parametrised by vectors $\mathbf{r}_\alpha(s), \mathbf{r}_\beta(s'), s, s' = 1, \dots, N$, and belonging to a topological class fully specified by a set of variables $m, p, q \dots$; here m is the value of the first invariant $I_{\alpha\beta}$ etc[†]. Fundamental

[†] Mathematically it has of course never been proved that any complete sequence of invariants exists; however, naively $I_{\alpha\beta}$ is of primary importance, for if $m \neq 0$ the loops are certainly knotted non-trivially, whilst other invariants have certainly been identified, so our notation at least allows us to clarify the nature of some preliminary estimates of the effects of topological entanglements.

quantities of interest are the total number of configurations $C_{mpq\dots}$ and averages such as the mean square size of a polymer loop (see figure 2):

$$\langle R^2 \rangle_{mpq\dots} = \frac{\sum_{s=1}^N \int dR^d R^2 C_{mpq\dots}(\mathbf{R}, s)}{NC_{mpq\dots}} \tag{2.2}$$

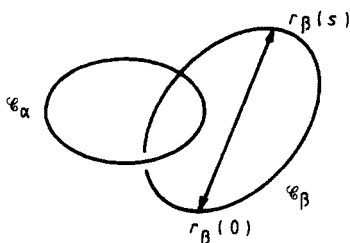


Figure 2.

Here $C_{mpq\dots}(\mathbf{R}, s)$ is the number of topologically equivalent configurations for which $r_\alpha(s) - r_\alpha(0) = \mathbf{R}$ and $I_{\alpha\beta} = m, \dots$

Naturally the calculation of $C_{mpq\dots}$, $\langle R^2 \rangle_{mpq\dots}$ demands further knowledge of the topological structure; however, we may obtain some useful information by comparing the results obtained for a polymer system when all the topological aspects are naively ignored with those for a system in which only the first invariant $I_{\alpha\beta}$ is respected (fixed)†.

Defining C_m to be the number of configurations for which the first invariant $I_{\alpha\beta}$ takes the value m and $C \equiv \sum_m C_m$ to be the overall total, then the reader will appreciate that the entropy $S_{mpq\dots}$ of the physical polymer system satisfies the inequality

$$S_{mpq\dots} \equiv \ln C_{mpq\dots} < S_m \equiv \ln C_m < S \equiv \ln C, \tag{2.3}$$

whence a valuable lower bound on the reduction in the entropy $\Delta S_{mpq\dots} = S_{mpq\dots} - S$ due to topological entanglements may be obtained.

To construct a representative average is more difficult, for knowing only the value m of the first invariant $I_{\alpha\beta}$, we must choose physically reasonable values for the remaining invariants. In the absence of further information, a suitable choice may be made by maximising $C_{mpq\dots}$ at fixed m , a condition which, if we weaken to allow a distribution of values $p, q, r \dots$ with probability g ,

$$g(p, q, r \dots | m) \equiv C_{mpq\dots} / C_m, \tag{2.4}$$

leads us to examine the quenched average $\overline{\langle R^2 \rangle}_m$ defined as follows for general distributions g :

$$\overline{\langle R^2 \rangle}_m = \sum_{p,q,r\dots} g(p, q, r \dots | m) \langle R^2 \rangle_{mpq\dots} \tag{2.5}$$

Now for the choice (2.4) the quenched average $\overline{\langle R^2 \rangle}_m$ coincides exactly with the mean square size of a polymer loop $\langle R^2 \rangle_m$ in an unphysical system for which only the first invariant $I_{\alpha\beta}$ is respected, for (2.2), (2.4) and (2.5) imply that

$$\langle R^2 \rangle_m \equiv \frac{1}{NC_m} \sum_{s=1}^N \int dR^d R^2 C_m(\mathbf{R}, s) = \overline{\langle R^2 \rangle}_m. \tag{2.6}$$

† The dynamics of such a system will be a little strange, for strictly we must allow all configurations for fixed m to mix, i.e. $2(a) \rightarrow 2(d) \not\rightarrow (2b)$, say.

Here $C_m(\mathbf{R}, s)$ is the number of configurations for which $\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) = \mathbf{R}$ and $I_{\alpha\beta} = m$. On this basis we may argue that a direct comparison between $\langle R^2 \rangle_m$, (2.6), and the naive expression

$$\langle R^2 \rangle = \frac{1}{NC} \sum_{s=1}^N \int d\mathbf{R}^d R^2 C(\mathbf{R}, s), \tag{2.7}$$

where $C(\mathbf{R}, s)$ is the number of configurations for which $\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) = \mathbf{R}$, should be a reliable guide to the effect of entanglements on the mean square size of a polymer $\langle R^2 \rangle_{mpq\dots}$. We should finally remark that, in contrast, the quenched entropy \bar{S}_m bears no simple relation to the entropy S_m , (2.3), even for the choice (2.4).

3. The Brereton–Shah problem

Following BS, we shall study the problem of two polymer loops topologically linked ($I_{\alpha\beta}$ respected) in the limit that one of the chains is allowed to fill a macroscopic volume at finite density $\bar{\rho}$, for this system may be reformulated in terms of an $n \rightarrow 0$ locally gauge-invariant field theory ideally suitable for a discussion of the effects of entanglements on universality.

Suppose we have two polymer loops $\mathcal{C}_\alpha, \mathcal{C}_\beta$ parametrised by vectors $\mathbf{r}_\alpha(s), \mathbf{r}_\beta(s')$ (see figure 2); then we may in terms of the simple phenomenological Hamiltonian H ,

$$\begin{aligned} H = & \int_0^{N_\alpha} ds_\alpha \left(\frac{\partial \mathbf{r}_\alpha(s_\alpha)}{\partial s_\alpha} \right)^2 + \frac{u}{4} \int_0^{N_\alpha} ds_\alpha \int_0^{N_\alpha} ds'_\alpha \delta[\mathbf{r}_\alpha(s_\alpha) - \mathbf{r}_\alpha(s'_\alpha)] \\ & + \frac{u}{2} \int_0^{N_\alpha} ds_\alpha \int_0^{N_\beta} ds_\beta \delta[\mathbf{r}_\alpha(s_\alpha) - \mathbf{r}_\beta(s_\beta)] \\ & + \int_0^{N_\beta} ds_\beta \left(\frac{\partial \mathbf{r}_\beta(s_\beta)}{\partial s_\beta} \right)^2 + \frac{u}{4} \int_0^{N_\beta} ds_\beta \int_0^{N_\beta} ds'_\beta \delta[\mathbf{r}_\beta(s_\beta) - \mathbf{r}_\beta(s'_\beta)], \end{aligned} \tag{3.1}$$

introduced by Edwards (1966) to describe the configurational statistics of polymers comprising $L_i = N_i A^2$ gaussian segments (links) $i = \alpha, \beta$ and interacting through the excluded volume coupling $u \Lambda^{-\epsilon}$ (dimensionless), write down the following expression for the number of configurations $C_m(\mathbf{R}, s)$ (cf (2.6) *et seq.*) for which $\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) = \mathbf{R}$ and $I_{\alpha\beta} = m$ (cf (2.1)):

$$C_m(\mathbf{R}, s) = \int [d\mathbf{r}_\alpha] \int [d\mathbf{r}_\beta] \delta(I_{\alpha\beta} - m) \delta[\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) - \mathbf{R}] \exp(-H). \tag{3.2}$$

Here $\int [d\mathbf{r}_i]$ represents a functional integral with the loop constraint $\mathbf{r}_i(N_i) = \mathbf{r}_i(0)$ and the scale Λ^{-1} is representative of the monomer spacing along the chain. Our discussion will centre around $C_m(\mathbf{R}, s)$, (3.2), for we may trivially construct the mean square size $\langle R^2 \rangle_m$ of the polymer loop \mathcal{C}_α or the entropy S_m in terms of this basic function as follows:

$$\langle R^2 \rangle_m \equiv \frac{1}{N_\alpha C_m} \int_0^{N_\alpha} ds \int d\mathbf{R}^d R^2 C_m(\mathbf{R}, s), \tag{3.3}$$

$$S_m \equiv \ln C_m, \tag{3.4}$$

$$C_m \equiv \frac{1}{N_\alpha} \int_0^{N_\alpha} ds \int dR^d C_m(\mathbf{R}, s)^\dagger. \tag{3.5}$$

Naturally, to compute the overall number of configurations $C(\mathbf{R}, s)$ such that $\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) = \mathbf{R}$ and thence $\langle R^2 \rangle$, s at (2.3), (2.7), we could use the identity

$$C(\mathbf{R}, s) = \sum_m C_m(\mathbf{R}, s). \tag{3.6}$$

However, it is instead more convenient simply to suppress the constraint $I_{\alpha\beta} = m$ in (3.2), yielding the expression

$$C(\mathbf{R}, s) = \int [d\mathbf{r}_\alpha] \int [d\mathbf{r}_\beta] \delta[\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) - \mathbf{R}] \exp(-H). \tag{3.7}$$

We are primarily interested in the limit that the polymer loop is allowed to fill a macroscopic volume at finite density $\bar{\rho}$, so it is useful to recast (3.2) and (3.7) in terms of a statistical average over the monomer density $\rho(\mathbf{R})$ and vector bond density $\mathbf{u}(\mathbf{R})$ associated with the polymer loop \mathcal{C}_β :

$$\rho(\mathbf{R}) = \int_0^{N_\beta} ds_\beta \delta[\mathbf{r}(s_\beta) - \mathbf{R}], \tag{3.8}$$

$$\mathbf{u}(\mathbf{R}) = \int_0^{N_\beta} d\mathbf{r}_\beta \delta[\mathbf{r}(s_\beta) - \mathbf{R}], \tag{3.9}$$

where we have included a rescaling by the monomer spacing Λ^{-1} in the definition

$$\bar{\rho} \equiv \frac{L_\beta}{V} = \Lambda^2 \tilde{\rho} \equiv \frac{\Lambda^2}{V} \int dR^d \rho(\mathbf{R}).$$

Formally integrating out the coordinates $\{\mathbf{r}_\beta\}$ of the background polymer loop \mathcal{C}_β in favour of the fields $\rho(\mathbf{R})$, $\mathbf{u}(\mathbf{R})$, we may rewrite (3.2) in the form

$$C_m(\mathbf{R}, s) = \int [d\mathbf{r}_\alpha] \int [d\mathbf{r}_\beta] \delta[\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) - \mathbf{R}] \exp(-\tilde{H}) \int [d\mathbf{u}] P \tag{3.10}$$

where the functionals \tilde{H} and P are given below:

$$\begin{aligned} \tilde{H} = & \int_0^{N_\alpha} ds \left(\frac{\partial \mathbf{r}_\alpha}{\partial s} \right)^2 + \frac{u}{4} \left(\int_0^{N_\alpha} ds \int_0^{N_\alpha} ds' \delta[\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(s')] \right. \\ & \left. + 2 \int ds \int dR^d \rho(\mathbf{R}) \delta(\mathbf{r}_\alpha - \mathbf{R}) + \int dR^d \rho^2(\mathbf{R}) \right), \end{aligned} \tag{3.11}$$

$$\begin{aligned} P = & \int [d\mathbf{r}_\beta] \delta(I_{\alpha\beta} - m) \prod_{\mathbf{R}} \delta \left(\mathbf{u}(\mathbf{R}) - \int_{\mathcal{C}_\beta} d\mathbf{r}_\beta \delta(\mathbf{r}_\beta - \mathbf{R}) \right) \delta \left(\rho(\mathbf{R}) - \int_{\mathcal{C}_\beta} ds_\beta \delta(\mathbf{r}_\beta - \mathbf{R}) \right) \\ & \times \exp \left[- \int_0^{N_\beta} ds_\beta \left(\frac{\partial \mathbf{r}_\beta(s_\beta)}{\partial s_\beta} \right)^2 \right]. \end{aligned} \tag{3.12}$$

Following Edwards (1967, 1968) and BS, we may simplify (3.12) and uncover the

† Here $\int dR^d C_m(\mathbf{R}, s)$ is independent of s and is averaged $\int_0^N ds/N$ purely for convenience.

underlying gauge symmetry of the problem by introducing a new field $\mathbf{A}(\mathbf{R})$:

$$\text{curl } \mathbf{A}(\mathbf{R}) = \mathbf{u}(\mathbf{R}), \tag{3.13}$$

$$\text{div } \mathbf{A}(\mathbf{R}) = 0. \tag{3.14}$$

Drawing on the direct analogy with electromagnetism, we see that most generally $\mathbf{A}(\mathbf{R})$ may be written as a line integral,

$$\mathbf{A}(\mathbf{R}) = \frac{1}{4\pi} \int_{\mathcal{C}_\beta} d\mathbf{r}_\beta \times \nabla \left(\frac{1}{|\mathbf{r}_\beta - \mathbf{R}|} \right), \tag{3.15}$$

so that the invariant $I_{\alpha\beta}$, (2.1), has a particularly simple representation in terms of $\mathbf{A}(\mathbf{R})$:

$$I_{\alpha\beta} = \int_{\mathcal{C}_\alpha} d\mathbf{r}_\alpha \cdot \mathbf{A}(\mathbf{r}_\alpha). \tag{3.16}$$

Guided by the simple structure of $I_{\alpha\beta}$, (3.16), we formally integrate out the coordinates $\mathbf{u}(\mathbf{R})$ in favour of the ‘vector potential’ $\mathbf{A}(\mathbf{R})$, rewriting (3.10) in the form

$$\begin{aligned} C_m(\mathbf{R}, s) = & \int [d\mathbf{r}_\alpha] \int [d\rho] \int [d\mathbf{A}] \prod_r \delta[\text{div } \mathbf{A}(r)] \\ & \times \delta \left(\int_{\mathcal{C}_\alpha} d\mathbf{r}_\alpha \cdot \mathbf{A} - m \right) \delta[\mathbf{r}_\alpha(s) - \mathbf{r}_\alpha(0) - \mathbf{R}] \exp(-\tilde{H}) \int [d\mathbf{u}] Q. \end{aligned} \tag{3.17}$$

Here the functional Q which describes the effect of the background polymer loop \mathcal{C}_β has the following structure:

$$\begin{aligned} Q = & \int d\mathbf{r}_\beta \prod_R \delta(\text{curl } \mathbf{A}(\mathbf{R}) - \int_{\mathcal{C}_\beta} d\mathbf{r}_\beta \delta(\mathbf{r}_\beta - \mathbf{R}) \delta(\rho(\mathbf{R}) - \int_{\mathcal{C}_\beta} ds \delta(\mathbf{r}_\beta - \mathbf{R})) \\ & \times \exp \left[- \int_0^{N_\beta} ds_\beta \left(\frac{\partial \mathbf{r}_\beta}{\partial s_\beta} \right)^2 \right]. \end{aligned} \tag{3.18}$$

In order to rewrite (3.18) in a useful form we must examine the physics of the finite density limit for the chain \mathcal{C}_β . Now for a random walk we would estimate $\bar{\rho} \sim L^{-d/2} \Lambda^d$ ($\bar{\rho}$ decreases as $L_\beta \rightarrow \infty$), whence to achieve a finite density $\bar{\rho}$ the containing box must actively restrict the polymer \mathcal{C}_β so that the evaluation of (3.18) is non-trivial even for this simple gaussian measure. Qualitatively, however, the dominant (and universal) structure expected arises from the correlation of $\rho(x)$, $\mathbf{u}(x)$ over distances $1 \ll x \Lambda \ll L_\beta^{1/2}$ (at least), which may realistically be modelled in terms of an equivalent system of M independent gaussian chains of an average length \bar{L} , say, providing $x \Lambda \ll \bar{L}^{1/2}$ and $\bar{\rho} = M\bar{L}/V$. In this sense we may therefore express the ‘dominant’ correlations in the form

$$\langle \rho(\mathbf{R}) \rangle \equiv \frac{\int [d\mathbf{r}_\beta] \rho(\mathbf{R}) \exp[-\int_0^{N_\beta} ds_\beta (\partial \mathbf{r}_\beta / \partial s_\beta)^2]}{\int [d\mathbf{r}_\beta] \exp[-\int ds_\beta (\partial \mathbf{r}_\beta / \partial s_\beta)^2]} = \frac{N_\beta}{V} = \tilde{\rho} = \Lambda^{-2} \bar{\rho}, \tag{3.19}$$

$$\langle \rho(k) \rho(-k) \rangle = \int d\mathbf{R}^d \exp(i\mathbf{k} \cdot \mathbf{R}) \langle \rho(\mathbf{R}) \rho(0) \rangle L \tilde{\rho} / k^2 (1 + O(1/Lk^2)), \tag{3.20}$$

$$\langle \mathbf{u}(\mathbf{R}) \rangle = \mathbf{0}, \tag{3.21}$$

$$\langle u_i(\mathbf{R}) u_j(\mathbf{R}') \rangle = \delta_{ij} \tilde{\rho} \delta(\mathbf{R} - \mathbf{R}'), \tag{3.22}$$

$$\langle \rho(\mathbf{R}) \mathbf{u}(\mathbf{R}') \rangle = \mathbf{0}, \tag{3.23}$$

over distances $x\Lambda \ll \bar{L}^{1/2}$ despite the constraint $\int dR^d \langle u(\mathbf{R})f(\{\rho\}, \{u\}) \rangle$ for all functionals f which follows from the loop constraint $\mathbf{r}_\beta(N_\beta) = \mathbf{r}_\beta(0)$. Higher correlations will only modify the structure of the dominant terms in the critical semi-dilute regime defined by the inequalities $uN_\alpha\bar{\rho} \gg 1$, $u\bar{\rho} \ll 1$ ($d < 4$) where we shall find that the topological entanglements are entirely negligible, so for our purpose these corrections can be ignored†.

Evaluating Q via (3.19)–(3.23), we find the exponentiated form

$$Q = \exp\left(-\frac{1}{\bar{\rho}}\right) \left(\int -dR^d (\nabla\rho(\mathbf{R}))^2 + (\text{curl } \mathbf{A}(\mathbf{R}))^2 \right) + O((\nabla^2\rho)\rho^2, (\text{curl } \mathbf{A})^4) \tag{3.24}$$

where the derivative couplings of the first corrections to the gaussian approximation ensure that such terms are only of interest for $d < 4$ dimensions despite the ϕ^3 nature of the field $\rho(\mathbf{R})$.

Combining finally the results (3.11), (3.17), (3.24) and introducing an integral representation for the topological constraint, it is now straightforward to obtain the expression

$$C_m(\mathbf{R}, s) = \int de \exp(iem) \int d\mathbf{R} \int d\mathbf{A} \int d\rho \prod_{\mathbf{R}} \delta[\text{div } \mathbf{A}(\mathbf{R})] \delta[\mathbf{r}(s) - \mathbf{r}(0) - \mathbf{R}] \exp(-\mathcal{L}) \tag{3.25}$$

where the functional \mathcal{L} closely resembles the Lagrangian of a quantum mechanical particle moving around a loop in an electromagnetic field specified by a vector potential $\mathbf{A}(\mathbf{R})$,

$$\begin{aligned} \mathcal{L} = & \int_0^N ds \left[\left(\frac{\partial \mathbf{r}}{\partial s} \right)^2 - ie\mathbf{A}(\mathbf{r}) \frac{\partial \mathbf{r}}{\partial s} + u\rho(\mathbf{r}) \right] + \frac{u}{4} \int_0^N ds \int_0^N ds' \delta[\mathbf{r}(s) - \mathbf{r}(s')] \\ & + \frac{1}{\bar{\rho}} \int dR^d (\text{curl } \mathbf{A}(\mathbf{R}))^2 + \frac{1}{\bar{\rho}} \int dR^d [(\nabla\rho(\mathbf{R}))^2 + u\rho^2(\mathbf{R})]. \end{aligned} \tag{3.26}$$

Here the function $\mathbf{r} = \mathbf{r}(s)$ satisfies the loop constraint $\mathbf{r}(N) = \mathbf{r}(0)$. For long flexible polymers, $L = N\Lambda^2 \gg 1$, (3.26) should describe the universal properties which arise from the correlations over scales x , $1 \ll x\Lambda \leq L^{1/2} \ll \bar{L}^{1/2}$. Our approximation scheme breaks down only if distances $x\Lambda \geq L^{1/2}$ become important; however, in such a situation the value of the phenomenological Hamiltonian H must surely be reviewed, so we shall assume from here that (3.26) is an adequate generalisation of the Edwards model (3.1) to the case of entanglements. More generally, (3.26) is of interest in its own right, for under the renormalisation group the couplings $e^2\bar{\rho}$, \bar{u} grow uncontrollably in $d < 4$ dimensions (see § 5), which would suggest that the representation is inadequate; in § 5 we shall show that this fear is unfounded for winding numbers $m \ll m_c$, \tilde{m}_c (5.37; $m_c, \tilde{m}_c \sim L^\alpha$, $\alpha > 0$).

In order to compare (3.25) and (3.26) with the BS expressions it is convenient to follow Edwards (1975) and integrate over the density field $\rho(\mathbf{R})$ to obtain

$$C_m(\mathbf{R}, s) = \int de \exp(iem) \int [d\mathbf{r}] \int [d\mathbf{A}] \prod_{\mathbf{R}} \delta[\text{div } \mathbf{A}(\mathbf{R})] \delta[\mathbf{r}(s) - \mathbf{r}(0) - \mathbf{R}] \exp(-\tilde{\mathcal{L}}) \tag{3.27}$$

† In fact this gaussian scheme describes the dominant terms in the dilute and semi-dilute limits correct to first order in the usual loop expansion (no entanglements).

where

$$\tilde{L} = \int_0^N ds \left(\frac{\partial \mathbf{r}}{\partial s} \right)^2 - ie\mathbf{A} \cdot \frac{\partial \mathbf{r}}{\partial s} + \frac{u}{4} \int_0^N ds \int_0^{N'} ds' \int d\mathbf{q}^d f(q) \exp\{iq[\mathbf{r}(s) - \mathbf{r}(s')]\} + \frac{1}{\tilde{\rho}} \int d\mathbf{R}^d (\text{curl } \mathbf{A}(\mathbf{R}))^2 \tag{3.28}$$

and

$$f(q) = q^2 / (q^2 + \tilde{\rho}u). \tag{3.29}$$

For polymer loops described by random flight statistics ($u = 0$), indeed (3.27), (3.28), (3.29) reduce to the expressions of BS; however, for $u \neq 0$ the ‘effective’ excluded volume $f(q)$ is a subtle function of the momentum transfer q , (3.3), so that the BS extension to loops with excluded volume is strictly only valid in the dilute $u\tilde{\rho}N \ll 1$ limit (see § 4).

To discuss the field theoretical approach to $\langle R^2 \rangle_m$, S_m it is simplest to restrict our attention to the derived function $C_m(\mathbf{R})$,

$$C_m(\mathbf{R}) \equiv \frac{1}{N} \int ds C_m(\mathbf{R}, s). \tag{3.30}$$

Proceeding by direct analogy with the linear polymer approach (Emery 1975), we first introduce the representation

$$\exp\left(-\frac{u}{4} \int_0^N ds \int_0^N ds' \delta[\mathbf{r}(s) - \mathbf{r}(s')]\right) = \int [d\psi] \exp\left[-\int d\mathbf{R}^d \left(\frac{1}{u} \psi(\mathbf{R})^2 + i \int_0^N ds \psi(\mathbf{R}) \delta[\mathbf{r}(s) - \mathbf{R}]\right)\right] \tag{3.31}$$

for the excluded volume term (cf (3.1)) in order to rewrite (3.30) in a factorised form. Explicitly we may express $C_m(\mathbf{R})$ via (3.30), (3.31) and (3.26) as follows:

$$C_m(\mathbf{R}) = \frac{1}{N} \int_0^N ds \int d\mathbf{e} \exp(iem) \int [d\psi] \int [d\mathbf{A}] \int [d\rho] \prod_R \delta[\text{div } \mathbf{A}(\mathbf{R})] \exp(-M) \times K(\mathbf{R}, s, \mathbf{A}) K(\mathbf{R}, s, -\mathbf{A}) \tag{3.32}$$

where the functionals $K(\mathbf{R}, s, [\mathbf{A}])$ and $M(\{\mathbf{A}\}, \{\rho\}\{\psi\})$ are of the form

$$K = \int [d\mathbf{r}] \delta[\mathbf{r}(s) - \mathbf{r}(0) - \mathbf{R}] \exp\left\{-\int_0^N ds \left[\left(\frac{\partial \mathbf{r}}{\partial s}\right)^2 - ie\mathbf{A} \frac{\partial \mathbf{r}}{\partial s} + \rho(r) + i\psi(r) \right]\right\}, \tag{3.33}$$

$$M = \int d\mathbf{R}^d \left(\frac{1}{u} \psi^2(\mathbf{R}) + \frac{1}{\tilde{\rho}} (\nabla \rho(\mathbf{R}))^2 + u\rho^2(\mathbf{R}) + \frac{1}{\tilde{\rho}} (\text{curl } \mathbf{A}(\mathbf{R}))^2 \right).$$

Laplace transforming (3.32) with respect to N then allows us to redevelop (3.32) in terms of the transformed functions as a simple product:

$$L_t(NC_m(\mathbf{R})) \equiv \int dN e^{-Nt} NC_m(\mathbf{R}) = \int d\mathbf{e} \exp(iem) \int [d\mathbf{A}] \int [d\rho] \int [d\psi] \exp(-M) \times K(\mathbf{R}, t, \mathbf{A}) K(\mathbf{R}, t, -\mathbf{A}), \tag{3.34}$$

$$L_t(K(\mathbf{R}, s, \mathbf{A})) \equiv \int ds e^{-st} K(\mathbf{R}, s, \mathbf{A}) \equiv K(\mathbf{R}, t, \mathbf{A}). \quad (3.35)$$

Now it is well known (see for example Feynman and Hibbs (1965)) that the function $K(\mathbf{R}, s, \mathbf{A})$ defined by equation (3.33) satisfies the differential equation

$$[\partial/\partial s - (\nabla - ie\mathbf{A})^2 + \rho + i\psi]K(\mathbf{R}, s, \mathbf{A}) = \delta(\mathbf{R}) \quad (3.36)$$

or equivalently

$$[t - (\nabla - ie\mathbf{A})^2 + \rho + i\psi]K(\mathbf{R}, t, \mathbf{A}) = \delta(\mathbf{R}) \quad (3.37)$$

so that we are led to make the formal identification

$$K(\mathbf{R}, t, \mathbf{A}) \stackrel{\text{no sum on } \alpha}{=} \langle \phi_\alpha(\mathbf{R})\phi_\alpha^*(0) \rangle = \frac{\int [d\phi] \phi_\alpha(\mathbf{R})\phi_\alpha(0) \exp[-H(\{\phi\})]}{\int d\phi \exp[-H(\{\phi\})]} \quad (3.38)$$

where the field theoretic average is computed from the following Hamiltonian:

$$H(\{\phi\}) = -\frac{1}{2} \sum_{\alpha=1}^n \int d\mathbf{R}^d \phi_\alpha^*(\mathbf{R}) [(\nabla - ie\mathbf{A})^2 + \rho + i\psi - t] \phi_\alpha(\mathbf{R}). \quad (3.39)$$

Here $\phi_\alpha(\mathbf{R})$ is an n -dimensional complex field. Furthermore, with the constraint $\nabla \cdot \mathbf{A} = 0$, (3.14), one may derive the complementary result

$$\mathbf{K}(\mathbf{R}, t, -\mathbf{A}) = \langle \phi_\alpha^*(\mathbf{R})\phi_\alpha(0) \rangle. \quad (3.40)$$

Using (3.38) and (3.40), we may then replace (3.34) by the form

$$L_t(NC_m(\mathbf{R})) = \int de \exp(iem) \int [d\mathbf{A}] \int [d\rho] \int [d\psi] \exp(-M) \times \langle \phi_\alpha(\mathbf{R})\phi_\alpha(0) \rangle \langle \phi_\beta^*(\mathbf{R})\phi_\beta(0) \rangle \quad (\text{no sum on } \alpha, \beta). \quad (3.41)$$

Specialising now to the $n \rightarrow 0$ limit of the field theory (3.39), we see that

$$Z \equiv \int d\phi \exp[-H(\{\phi\})] = 1 \quad (3.42)$$

whilst the gaussian nature of (3.39) ensures that

$$\langle \phi_\alpha(\mathbf{R})\phi_\alpha^*(0)\phi_\beta(\mathbf{R})\phi_\beta(0) \rangle \stackrel{\alpha \neq \beta}{=} \langle \phi_\alpha(\mathbf{R})\phi_\beta^*(0) \rangle \langle \phi_\beta^*(\mathbf{R})\phi_\beta(0) \rangle \quad (3.43)$$

so that we may reformulate (3.41) as follows:

$$L_t(NC_m(\mathbf{R})) = \lim_{n \rightarrow 0} \int de \exp(iem) \int [d\mathbf{A}] \int [d\rho] \int d\psi \exp(-M) \times \int [d\phi] \phi_\alpha(\mathbf{R})\phi_\alpha^*(0)\phi_\beta^*(\mathbf{R})\phi_\beta(0) \exp[-H(\{\phi\})], \quad (3.44)$$

whence, finally integrating out the field $\psi(\mathbf{R})$, (3.31), (3.33), we obtain the result

$$C_m(\mathbf{R}) = \frac{1}{N} \lim_{n \rightarrow 0} \int de \exp(iem) L_t^{-1} \langle \phi_\alpha(\mathbf{R})\phi_\alpha^*(0)\phi_\beta^*(\mathbf{R})\phi_\beta(0) \rangle \quad (3.45)$$

where the field theoretic average is computed for the Hamiltonian of three fields $\phi_\alpha(\mathbf{R})$,

$\rho(\mathbf{R}), \mathbf{A}(\mathbf{R})$:

$$H \equiv \int d\mathbf{R}^d \left[\frac{1}{\tilde{\rho}} (\text{curl } \mathbf{A})^2 + \frac{1}{2} \sum_{\alpha=1}^n |(\nabla - ie\mathbf{A})\phi_\alpha|^2 + t\phi_\alpha^2 + u\rho|\phi_\alpha|^2 + \frac{u}{4} \left(\sum_{\alpha=1}^n |\phi_\alpha|^2 \right)^2 + \frac{1}{\tilde{\rho}} (\nabla\rho)^2 + u\rho^2 \right]. \tag{3.46}$$

Here L_t^{-1} denotes the operation of Laplace inversion. Alternatively we can, at the cost of introducing a non-local ϕ^4 coupling, integrate out the $\rho(\mathbf{R})$ field exactly to leave an effective two-field Hamiltonian \tilde{H} (cf (3.28), (3.29)),

$$\tilde{H} \equiv \int d\mathbf{R}^d \left(\frac{1}{\tilde{\rho}} (\text{curl } \mathbf{A})^2 + \frac{1}{2} \sum_{\alpha=1}^n |(\nabla - ie\mathbf{A})\phi_\alpha|^2 + t\phi_\alpha^2 \right) + \frac{u}{4} \sum_{\alpha,\beta=1}^n \int d\mathbf{R}^d \int d\mathbf{R}'^d \int dq^d \exp[iq(\mathbf{R} - \mathbf{R}')f(q)] |\phi_\alpha(\mathbf{R})|^2 |\phi_\beta(\mathbf{R}')|^2 \tag{3.47}$$

where as before the effective potential $f(q)$ is given by equation (3.29).

Our results (3.46) or (3.47) represent the correct generalisation of the BS expressions valid for polymer loops obeying random flight statistics. As remarked before, cf (3.27) *et seq.*, in the dilute regime $u\tilde{\rho}N \ll 1$, $f(q) \rightarrow 1$, leaving a local effective Hamiltonian H^* ,

$$H^* \equiv \int d\mathbf{R}^d \left(\frac{1}{\tilde{\rho}} (\text{curl } \mathbf{A})^2 + \frac{1}{2} \sum_{\alpha=1}^n |(\nabla - ie\mathbf{A})\phi_\alpha|^2 + t|\phi_\alpha|^2 \right) + \frac{u}{4} \left(\sum_{\alpha=1}^n |\phi_\alpha|^2 \right)^2, \tag{3.48}$$

which may be compared directly with the result conjectured by BS for loops with excluded volume interactions. BS argue that the effective Hamiltonian is of a form similar to Hertz (1978),

$$H^{**} \equiv \int d\mathbf{R}^d \left(\frac{1}{\tilde{\rho}} (\text{curl } \mathbf{A})^2 + \frac{1}{2} \sum_{\alpha=1}^n |(\nabla - ie\mathbf{A})\phi_\alpha|^2 + t|\phi_\alpha|^2 + \frac{u}{4} |\phi_\alpha|^4 \right), \tag{3.49}$$

which in contrast with (3.48), ... possesses an $S_n \times O(2)$ rather than $O(2n)$ global symmetry. Here S_n is the permutation group of n objects. Physically BS are surely in error, for in the limit $\tilde{\rho} \rightarrow 0$ and $m = 0$ we would hope that the topological constraint is irrelevant. This is indeed the situation, for $\tilde{\rho} \rightarrow 0$ implies $\text{curl } \mathbf{A} = 0$, cf (3.48), and thus $\mathbf{A} = \nabla\theta(\mathbf{R})$ for some scalar potential $\theta(\mathbf{R})$, allowing us to eliminate the topological constraint by means of the local gauge transformation

$$\begin{aligned} \mathbf{A} &\rightarrow \mathbf{A}' + \nabla\theta(\mathbf{R}) \\ \phi_\alpha &\rightarrow e^{i\theta(\mathbf{R})} \phi'_\alpha \end{aligned} \quad (\mathbf{A} \equiv 0, \tilde{\rho} \rightarrow 0) \tag{3.50}$$

to leave the canonical $O(p)$ symmetric field theoretic representation for the statistics of single polymer chains

$$H = \int d\mathbf{R}^d \frac{1}{2} \sum_{\alpha=1}^n [(\nabla\phi_\alpha)^2 + t\phi_\alpha^2] + \frac{u}{4} \left(\sum_{\alpha=1}^n \phi_\alpha^2 \right)^2 \tag{3.51}$$

(Elderfield 1978, des Cloizeaux 1975). For our purposes this discrepancy is essential, for the Hertz model contains a further renormalisation group instability which cannot be suppressed by the mechanism employed in §§ 4 and 5 for the entanglement problem.

4. The random flight ($u\Lambda^{-\epsilon} \ll 1$) limit of the Brereton–Shah problem

Conventionally as a first move theoreticians have for many years factored out the non-universal features of the polymer functions of interest, presenting them as simple scale factors. In contrast, we shall argue that in the presence of topological constraints there is an important coupling between the universal and non-universal aspects of the problem which leads via a subtle mechanism to the conclusion that entanglements are irrelevant in the long-chain limit. We shall approach the problem in this section by showing that entanglements modify functions characterising the large-scale (or universal) behaviour by terms typically of order $1/L$ in the perturbative $u\Lambda^{-\epsilon} \ll 1$ or random flight limit of the excluded volume statistics imposed on the polymer chains.

Our discussion focuses first on the function $C_m(R, N)$ which may be computed via the field theoretic connection (3.45),

$$C_m(R) = \frac{\alpha \neq \beta}{N} \lim_{n \rightarrow 0} \int de \exp(iem) L_i^{-1} \langle \phi_\alpha(R) \phi_\alpha^*(0) \phi_\beta^*(R) \phi_\beta(0) \rangle, \tag{4.1}$$

where the field theoretic average is computed using the Hamiltonian (3.47):

$$H = \int dx^d \left(\frac{1}{\tilde{\rho}} (\text{curl } \mathbf{A})^2 + \frac{1}{2} \sum_{\alpha=1}^n |(\nabla - i\mathbf{e}\mathbf{A})\phi_\alpha(x)|^2 + t|\phi_\alpha|^2 \right) + \frac{u}{4} \sum_{\alpha, \beta}^n \int dx^d dx'^d \int dq^d f(q) \exp[iq(x - x')] |\phi_\alpha(x)|^2 |\phi_\beta(x')|^2. \tag{4.2}$$

To investigate (4.1) we employ the usual perturbative scheme, which may be expressed in terms of Feynman graphs with propagators

$$\langle A_\mu(q) A_\nu(-q) \rangle \equiv q^{-2} (\delta_{\mu\nu} - q_\mu q_\nu q^{-2}) \equiv \text{wavy line},$$

$$\langle \phi_\alpha(q) \phi_\beta(-q) \rangle \equiv \delta_{\alpha\beta} / (q^2 + t) \equiv \text{solid line},$$

in momentum space and vertices

$$\text{wavy line} = -(e^2 \tilde{\rho}),$$

$$\text{solid line} = i(p_\mu + \frac{1}{2}q_\mu) \sqrt{e^2 \tilde{\rho}},$$

$$\text{cross} = +u.$$

Although our entanglement problem was formulated in $d = 3$ spatial dimensions, it is fruitful to consider the continuation of this perturbation theory to d dimensions. Of course for $d = 2$ or $d > 3$ the real entanglement problem becomes trivial (see § 1).

Simple power counting arguments (see Brézin *et al* 1973 (BLZ)) pick out $d = 4$ as the borderline dimension below which the dimensionless couplings $u\Lambda^{-\epsilon}$, $e^2 \tilde{\rho} \Lambda^{-\epsilon}$ ($d = 4 - \epsilon$) become relevant to the physics of the critical domain $R\Lambda \gg 1$, $t/\Lambda^2 \ll 1$ or equivalently $L = N\Lambda^2 \gg 1$, where we expect to observe universality. For $d > 4$ the expansion becomes trivial in the sense that the zeroth-order result

$$C_m(R) \equiv A \int_0^N dx \left(\frac{1}{N+x} \right)^{d/2} \exp\left[-\left(\frac{R^2}{N-x}\right)\right] \frac{1}{x^{d/2}} \exp\left(-\frac{R^2}{x}\right), \tag{4.3}$$

$A = 1,$

is modified only by a change in the amplitude $A = A(e^2 \tilde{\rho} \Lambda^{-\epsilon}, u \Lambda^{-\epsilon})$ independent of R, L in the critical domain, so that as expected ‘entanglements’ do not modify the essential physics for $d > 4$ within this simple continuation. In contrast, for $d < 4$ perturbatively the couplings are strongly relevant, for deep in the critical regime the expansions fail through the growth of the appropriate dimensionless couplings $uN^{\epsilon/2}, e^2 \tilde{\rho} N^{\epsilon/2}, \dots$. In order to control these expansions we shall seek in § 5 a renormalisation group scheme in $d = 4 - \epsilon$ ($\tilde{u} \equiv u \Lambda^{-\epsilon}, \tilde{x} = e^2 \tilde{\rho} \Lambda^{-\epsilon}; \tilde{x}, \tilde{u} = O(\epsilon)$); however, as a preliminary we first examine the constraints imposed on the polymer functions $C_m(R, N)$ in the perturbative or free chain domain $u \Lambda^{-\epsilon} \ll 1$ by the constraint that the topological entanglements be irrelevant.

In the critical domain the large length scales $N^{1/2}, (u\tilde{\rho})^{-1/2}$ dominate the physics and we expect to observe universal behaviour in terms of some renormalised couplings which précis the effects of the short-range ($\sim \Lambda^{-1}$) or non-universal aspects of the system. Following BLZ, we may rephrase $C_m(R, N)$ in terms of variables appropriate to the critical regime by employing a renormalisation scheme for $d \leq 4$ ($f(q) \rightarrow 1, q \rightarrow \infty$) dimensions and in particular the physical dimension $d = 3$ of primary interest. Specialising first to $d < 4$, this programme may be accomplished by a simple shift in the temperature t ,

$$t = \tilde{t} - \Delta \Lambda^2, \tag{4.4}$$

where we choose Δ (dimensionless) such that $\tilde{t} = 0$ locates the critical point at which the dominant correlation length diverges, or equivalently where the inverse susceptibility χ^{-1} vanishes. As usual the ‘magnetic’ susceptibility χ is directly related to the two-point Green function $G^2(q, t)$ taken at zero momentum,

$$G_{\alpha\beta}^2(q, t) \equiv \int dR^d \exp(iq) R \langle \phi_\alpha(R) \phi_\beta(0) \rangle,$$

$$\delta_{\alpha\beta} \chi \equiv G_{\alpha\beta}^2(0, t),$$

so evaluating the constraint $\chi^{-1}(\tilde{t}) = 0$ at $\tilde{t} = 0$ we find perturbatively

$$\Delta(e^2 \tilde{\rho} \Lambda^{-\epsilon}, u \Lambda^{-\epsilon}) = \text{[diagram 1]} + \text{[diagram 2]} + O((e^2 \tilde{\rho})^2, u^2) \tag{4.5}$$

where at least to first order in $e^2, \partial\Delta/\partial e^2 > 0$. After this renormalisation and transformation to momentum space we may rewrite $C_m(q, N)$ in the form

$$C_m(q, N) = N^{-d/2} \int de \exp(iem - \Delta L) G \tag{4.6}$$

where by construction the dominant contribution \bar{C}_m in the critical domain may be expressed as follows in terms of a scaling function \bar{G} which, depending only on the macroscopic scales $N^{1/2}, (u\tilde{\rho})^{-1/2}$, is insensitive to the short-distance (Λ^{-1}) structure of the chains:

$$\bar{C}_m(q, N) = N^{-d/2} \int de \exp(iem - \Delta L) \bar{G}(q^2 N, uN^{\epsilon/2}, e^2 \tilde{\rho} N^{\epsilon/2}, u\tilde{\rho} N). \tag{4.7}$$

Typically to isolate \bar{G} we may place the system deep into the critical domain $q^2, 1/N, u\tilde{\rho} \ll \Lambda^2$ by taking the limit $\Lambda \rightarrow \infty$ such that

$$\bar{G} \equiv \lim_{\Lambda \rightarrow \infty} G \tag{4.8}$$

at fixed $q, N, u\tilde{\rho}, e^2, u$. Rephrased in these terms, we therefore see that the universality exhibited by $C_m(q, N)$ through G will generally be modified by the essentially non-universal function $\Delta = \Delta(e^2\tilde{\rho}\Lambda^{-\varepsilon}, u\Lambda^{-\varepsilon})$ which depends strongly on the small-scale details of the underlying polymers. Here $G(q, N)$ is of course directly related to the Green function $\langle\phi\phi\phi\rangle$ via (4.1), (4.4) as follows:

$$N^{d/2}G(q, N) \stackrel{\alpha \neq \beta}{=} \frac{1}{N} \lim_{n \rightarrow 0} L_i^{-1} \int dR^d \exp(iq \cdot R) \langle \phi_\alpha(R) \phi_\alpha^*(0) \phi_\beta^*(R) \phi_\beta(0) \rangle. \quad (4.9)$$

Analysing first the entropy $S_m = \ln C_m$, (2.3), we see that for long chains $L \gg 1$ (or critical physics) the dominant contribution is of the form

$$\exp(S_m) = N^{-d/2} \int de \exp(iem - \Delta L) \bar{G}(q, N)|_{q=0} \quad (4.10)$$

where $\bar{G}(q, N)$ is given graphically by the expression†

$$L_i(N^{-d/2}G(q, N)) \stackrel{d < 4}{=} \lim_{\Lambda \rightarrow \infty} [\text{Diagram 1} + \text{Diagram 2} + \text{Diagram 3} + O((e^2\tilde{\rho})^2, u^2)].$$

To simplify the discussion it is useful to examine the physically realisable limits of dilute $u\tilde{\rho}N \ll 1$ and semi-dilute $u\tilde{\rho}N \gg 1$ physics separately, where $o_v \equiv u\tilde{\rho}N$ is a direct measure of the overlap between the chains $\mathcal{C}_\alpha, \mathcal{C}_\beta$. For dilute physics we find by proceeding to the extreme limit $\tilde{\rho} = 0$ ($e^2\tilde{\rho} \neq 0$) that the dominant contributions to $G(q)$ at $q = 0$ are of the form

$$\bar{G}(q)|_{q=0} \cong \sum_{k,p=0}^{\infty} a_{kp} (e^2\tilde{\rho})^k u^p N^{(k+p)\varepsilon/2} \quad (4.11)$$

where the coefficients a_{kp} are independent of $\tilde{\rho}, N$ up to small corrections of $O(u\tilde{\rho}N)$. Perturbatively we thus observe that as the relevant length scale $N^{1/2}$ grows, the expansion fails through the growth of the dimensionless couplings $uN^{\varepsilon/2}, e^2\tilde{\rho}N^{\varepsilon/2}$. Similarly, by taking the extreme limit $N \rightarrow \infty$ (or $\tilde{t} \rightarrow 0$) as necessary, we find that deep in the semi-dilute regime $u\tilde{\rho}N \gg 1$

$$\bar{G}(q)|_{q=0} \cong 1 + \sum_{k=1}^{\infty} b_k (e^2\tilde{\rho})^k N^{k\varepsilon/2} + \sum_{k,p=1}^{\infty} c_{kp} (e^2\tilde{\rho})^k u^p (u\tilde{\rho})^{-(k+p)\varepsilon/2} \quad (4.12)$$

where we see that for the entanglement problem the semi-dilute domain is characterised by both the length scales $N^{1/2}, (u\tilde{\rho})^{-1/2}$, in contrast to the problem without topological constraints (say $e^2 = 0$) for which only the $(u\tilde{\rho})^{-1/2}$ is important.

Introducing (4.11) and (4.12) into (4.10), the reader will observe that at least for d near four ($\varepsilon \ll 1$) the coefficient of the leading exponential will not be modified in the limit $L \gg 1$ of polymer physics, and therefore, in order to extract the dominant term, we may evaluate the integral over the charge ‘ e ’ by saddle point methods ($L \gg 1, \tilde{\rho} \neq 0, m$ fixed). In this way we may systematically obtain corrections to the mean field (or $d > 4$) result through which we may make predictions for the physical dimension $d = 3$ of interest.

We are thus led to seek solutions e^* of the saddle point equation

$$\partial\Delta/\partial e^2 = 0$$

† Rather than compute the integrals for fixed Λ , it is useful in practice to take $\Lambda = \infty$ at the outset and employ a dimensional regularisation scheme.

which are stable to small fluctuations

$$\partial^2 \Delta / (\partial e^2)^2 > 0 \quad \text{for } e = e^*$$

providing $L \gg 1$, m fixed or more carefully $m^2 \ll (L\tilde{\rho})^2$ as L increases. Now the function $\Delta(e^2 \tilde{\rho} \Lambda^{-\epsilon}, u \Lambda^{-\epsilon})$, (4.3), (4.5), is non-universal, for it depends on the small-scale (Λ^{-1}) structure, and furthermore it oscillates wildly so that naively our task appears to be entirely non-trivial. On physical grounds, however, we may argue that the origin $e^* = 0$ is a globally stable minimum which dominates the behaviour of the integral. We first remark that $e^* = 0$ is certainly a locally stable stationary point, (4.5). Now suppose a solution $e^* \neq 0$ is a global minimum of Δ ; then by definition

$$\Delta(e^*, u) < \Delta(0, u) \tag{4.13}$$

which implies that up to logarithmic corrections (2.3), (4.10)

$$S_m = -L\Delta(e^*, u) > -L\Delta(0, u). \tag{4.14}$$

In contrast, in the absence of topological constraints the entropy S is of the form

$$S = -L\Delta(0, u) + O(\ln L). \tag{4.15}$$

Therefore (4.14) is in complete contradiction with the physical expectation that topological constraints reduce the entropy $S_m < S$, (2.3), whence $e^* = 0$ is indeed a global minimum.

Assured now that the gaussian saddle point $e^* = 0$ dominates the behaviour of (4.10) up to exponentially small corrections $\sim \exp[-L(\Delta(e^*) - \Delta(0))]$ from the subsidiary solutions $e^* \neq 0$, we may carry out the gaussian integration to obtain via the identity

$$\int de \exp(2iem - e^2) f(e^2) = f\left(-\frac{\partial}{\partial \alpha}\right) \frac{1}{(\alpha\pi)^{1/2}} \exp\left(-\frac{m^2}{\alpha}\right) \Big|_{\alpha=1} \tag{4.16}$$

an expression for S_m factorised into universal and non-universal components. Summarising the universal or large-scale aspects by $\bar{G}(q^2, e^2 \tilde{\rho}, u, N)$ and the essential non-universal components in the long-chain limit $L \gg 1$ by $\Delta(e^2 \tilde{\rho}, u)$ and $\bar{\omega}(u)$,

$$\bar{\omega}(u) \equiv \frac{\partial}{\partial x} \Delta(x, u \Lambda^{-\epsilon}) \sim \Lambda^{-\epsilon} (1 + O(u \Lambda^{-\epsilon})), \quad x = e^2 \tilde{\rho}, \tag{4.17}$$

we may write the dominant contribution to S_m in the form (see (4.6) *et seq.*)

$$\begin{aligned} \exp(S_m) = & \bar{G}\left(q, -\frac{1}{L\bar{\omega}} \frac{\partial}{\partial \alpha}, u, N\right) \Big|_{q=0} \exp\left\{-\left[L\Delta\left(-\frac{1}{L\bar{\omega}} \frac{\partial}{\partial \alpha}, u\right) + \frac{\partial}{\partial \alpha}\right]\right\} \\ & \times \frac{1}{(\pi\alpha\tilde{\rho})^{1/2}} \exp\left(-\frac{m^2}{L\tilde{\rho}\bar{\omega}\alpha}\right) \Big|_{\alpha=1}. \end{aligned} \tag{4.18}$$

In order to judge the importance of topological entanglements in the universal domain $L \gg 1$, $u\tilde{\rho} \ll \Lambda^2$ of the naive solution

$$\exp(S) = \bar{G}(q, 0, u, N) \Big|_{q=0} \exp[-L\Delta(0, u)] \tag{4.19}$$

which may be obtained by ignoring the topological aspects entirely, we shall make a direct comparison between (4.19) and the factored form (4.18) valid when $I_{\alpha\beta}$ is respected and $L \gg 1$.

Analysing first the non-universal function Δ , we see from the identity

$$\left[\frac{1}{(L\tilde{\rho})^{1/2}\tilde{\omega}} \exp\left(-\frac{m^2}{L\tilde{\rho}\tilde{\omega}}\right) \right]^{-1} - \left(\frac{1}{L\tilde{\omega}} \frac{\partial}{\partial \alpha} \right)^k \left[\frac{1}{(L\tilde{\rho})^{1/2}\tilde{\omega}} \exp\left(-\frac{m^2}{L\tilde{\rho}\tilde{\omega}\alpha}\right) \right] \\ \simeq \left(\frac{1}{L\tilde{\omega}} \right)^k \left[1 + \dots \left(\frac{m^2}{L\tilde{\rho}\tilde{\omega}} \right)^k \right] \tag{4.20}$$

that order by order in $u\Lambda^{-\varepsilon}$ the corrections induced are of the form

$$L\tilde{\omega}_k \left(\frac{1}{L\tilde{\omega}} \right)^k \left[1 + \dots \left(\frac{m^2}{L\tilde{\rho}\tilde{\omega}} \right)^k \right], \quad k \geq 2, \tag{4.21}$$

where $\tilde{\omega}_k \equiv (\partial/\partial x^k)\Delta(x, u)$, $x = e^2\tilde{\rho}$. Observing that $\tilde{\omega}$, $\tilde{\omega}_k$ are independent of L , we see here that for long chains $L \gg 1$ topological entanglements are unimportant providing

$$m^2 \ll L^{2-1/k}\tilde{\rho}\tilde{\omega} \left(\frac{\tilde{\omega}}{(\tilde{\omega}_k)^{1/k}} \right) \ll L^{3/2}\tilde{\rho}\tilde{\omega} \left(\frac{\tilde{\omega}}{\sqrt{\omega_2}} \right) \tag{4.22}$$

where $(\tilde{\omega}_k)^{1/k}/\tilde{\omega}$, $\tilde{\omega}\Lambda^\varepsilon$, naively functions only of the dimensionless coupling $u\Lambda^{-\varepsilon}$, summarise the effect of the short-range or non-universal correlations and are therefore strongly model dependent. We take $(\tilde{\omega}_k)^{1/k}/\tilde{\omega}$, $\tilde{\omega}\Lambda^\varepsilon \sim 1$. With regard to the consistency of (4.22), it is important to notice that for our continuation the winding number has dimension $\Lambda^{1-\varepsilon}$, reducing only to a pure number for the physical entanglement problem ($d=3$ or $\varepsilon=1$). Interpreting (4.22), we first observe that deep in the semi-dilute regime for which the polymers \mathcal{C}_α , \mathcal{C}_β are strongly overlapping, $\sigma_v \equiv u\tilde{\rho}N \gg 1$ (see (4.12)), the constraint (4.22) is automatically satisfied for all finite m , so entanglements do not modify the physics described by Δ in this case. For the dilute limit $\sigma_v \equiv u\tilde{\rho}N \ll 1$ (see (4.11)), we find in direct contrast that (4.22) represents an important constraint on the system if $m \neq 0$, which reflects the difficulty of forming complex knots without affecting the large-scale properties of the system when the overlap is small; for through $\tilde{\omega}$, $\tilde{\omega}_k$ the chains are inflexible at scales $\sim \Lambda^{-1}$. Of course for $m=0$ the polymers can adopt an untangled configuration, since the higher-order invariants are not respected, so we find that (4.22) is irrelevant for all densities ($L \gg 1$) as one might expect.

For \tilde{G} we find modifications through (4.11), (4.12), (4.18), (4.21), which typically for dilute physics $u\tilde{\rho}N \ll 1$ are given by the estimates

$$\sim (1/L\tilde{\omega})^k [1 + \dots (m^2/L\tilde{\rho}\tilde{\omega})^k] N^{ke/2}, \quad k \geq 1, \tag{4.23}$$

which may be compared with the semi-dilute estimates applicable in the domain of strong overlap

$$\sim (1/L\tilde{\omega})^k [1 + \dots (m^2/L\tilde{\rho}\tilde{\omega})^k] N^{ke/2}$$

and

$$\sim u^p \left\{ \left(\frac{1}{L\tilde{\omega}} \right)^k \left[1 + \dots \left(\frac{m^2}{L\tilde{\rho}\tilde{\omega}} \right)^k \right] N^{ke/2} \right\} \left(\frac{1}{N\tilde{\rho}u} \right)^{ke/2} \left(\frac{1}{\tilde{\rho}u} \right)^{pe/2}. \tag{4.24}$$

Analogous to (4.22), we therefore find that entanglements are irrelevant to the physics described by \tilde{G} if the winding number m satisfies the constraint

$$m^2 \ll L^{d/2}\tilde{\rho}\tilde{\omega}^2. \tag{4.25}$$

As before we find that the induced constraint (4.25) is automatically satisfied if the

overlap between the polymers is large, whilst in the dilute regime it plays a significant role. It is worth remarking that within the perturbative framework $u\Lambda^{-\epsilon} \ll 1$, the size of a polymer chain $\sim L^{1/2}/\Lambda$ (random walk), so that (4.24) indeed places a bound on the overlap $\bar{\sigma}_v \equiv (L^{1/2}/\Lambda)^d \bar{\rho}$ below which entanglements (fixed m) will play a significant role. Here $\bar{\rho} \equiv \Lambda^2 \bar{\rho}$ (cf (3.8) *et seq.*) in the link density of the background chain \mathcal{C}_β .

Overall we see that for polymers obeying *random flight statistics* $u\Lambda^{-\epsilon} \ll 1$ entanglements are uninteresting when the polymers are strongly overlapping ($u\bar{\rho}N \gg 1$), whilst in the limit of small overlap ($u\bar{\rho}N \ll 1$) there are important constraints (4.22), (4.24),

$$m^2/\bar{\rho}\bar{w} \ll L^{3/2}, \quad d=3, L \gg 1, \quad (4.26)$$

which reflect the difficulty of forming complex ($m > 0$) knots without affecting the large-scale or universal behaviour of the chains when the overlap is small and the chains are inflexible at scales $\sim \Lambda^{-1}$. Qualitatively it is clear that \bar{w} is directly related to local stiffness of the chain, for suppose 'loops' of l_0 links $1 < l_0 \ll L$ are energetically unfavourable; then the model (3.1) overestimates the non-universal function \bar{w} by terms $\sim (1/l_0)(l_0/\Lambda^2)^{\epsilon/2}$. Assuming that the winding number m satisfies the overlap constraint (4.26), then we see immediately from (4.18), (4.19) that the entanglements reduce the entropy S by an amount $\bar{\Delta}$,

$$\bar{\Delta} \equiv S - S_m = \frac{m^2}{L\bar{\rho}\bar{w}} + \frac{1}{2} \ln(\pi L\bar{\rho}\bar{w}) \quad (4.27)$$

or equivalently that the configuration numbers C_m, C are related as follows:

$$C_m/C = (\pi L)^{-1/2} \exp(-m^2/L\bar{\rho}\bar{w})/\bar{\rho}\bar{w}. \quad (4.28)$$

Typically our approximation scheme is valid for $m^2 \ll L^{3/2}\bar{\rho}\bar{w}$, (4.26), so (4.28) can represent a massive reduction in the ratio C_m/C for long chains $L \gg 1$. The result is of particular interest, for if two polymers were originally formed together say in a melt, it is reasonable to estimate that $I_{\alpha\beta}$ will take the value m with probability C_m/C . In general, then, (4.26) will not be violated, for with high probability $m^2 \ll L\bar{\rho}\bar{w}$ so entanglements are relatively unimportant in determining say the mean square size of a chain $\langle R^2 \rangle$, for which a parallel argument shows that for both dilute and semi-dilute physics

$$\frac{\langle R^2 \rangle_m}{\langle R^2 \rangle} = \left[1 - \frac{\bar{C}}{L\bar{w}} \left(1 - \frac{m^2}{L\bar{\rho}\bar{w}} \right) L^{\epsilon/2} \right] \sim 1. \quad (4.29)$$

Here $\bar{C} > 0$ is a numerical coefficient and for simplicity we have suppressed the corrections arising from terms of $O(e^{2k})$, $k > 1$ (cf (5.31) *et seq.*). Of course if the two loops are formed separately the invariant $I_{\alpha\beta}$ takes the value $m = 0$, the constraint trivialises and we always find for long chains $L \gg 1$ the behaviour $\langle R^2 \rangle_m \sim \langle R^2 \rangle$, $C_m \sim C$. For $L \gg 1$ (4.29) has also been derived by the Brereton and Shah (1981) mean field techniques in the form

$$\langle R^2 \rangle_m / \langle R^2 \rangle = 1 - m^2/m^*_{\bar{C}} \quad (4.30)$$

where $m^*_{\bar{C}} \equiv \bar{C}L^{\alpha/2}\bar{\rho}\bar{w}^2$ and $\bar{C} > 0$.

The above results of course apply only in the domain of random flight statistics where $u\Lambda^{-\epsilon}$ is sufficiently small to ensure that even for $L \gg 1$ the excluded volume parameters $z \equiv uN^{\epsilon/2}$, $u(\bar{\rho}u)^{-\epsilon/2}$ relevant to the physics of the dilute and semi-dilute limits respectively do not modify the universal (or large-scale) structure.

5. The asymptotic behaviour of the Brereton–Shah model

We observed in § 4 that the dominant contribution $\bar{C}_m(q, N)$ to the function $C_m(q, N)$ of primary interest may be factored in the form

$$\bar{C}_m(q, N) = N^{-d/2} \int d\epsilon \exp(i\epsilon m - \Delta L) \bar{G}(q^2 N, e^2 \tilde{\rho} N^{\epsilon/2}, u N^{\epsilon/2}, u \tilde{\rho} N) \tag{5.1}$$

where $\Delta = \Delta(e^2 \tilde{\rho} \Lambda^{-\epsilon}, u \Lambda^{-\epsilon})$ is a non-universal function independent of the macroscopic scales $L^{1/2}/\Lambda \equiv N^{1/2}$, $u \tilde{\rho}^{-1/2}$ relevant respectively to the physics of the dilute and semi-dilute limits, and \bar{G} is in contrast a universal function depending strongly only on these macroscopic scales.

Focusing attention first on the dilute regime, we argued that for $o_v = u \tilde{\rho} N \ll 1$ (cf (4.11)) the relation (5.1) may be replaced by the simplified form

$$\bar{C}_m(q, N) = N^{-d/2} \int d\epsilon \exp(i\epsilon m - \Delta L) \bar{G}(q^2 N, e^2 \tilde{\rho} N^{\epsilon/2}, u N^{\epsilon/2}) \tag{5.2}$$

where $L^{1/2}/\Lambda \equiv N^{1/2}$ is the only relevant length scale and \bar{G} may be developed perturbatively in the dimensionless couplings $z \equiv u N^{\epsilon/2}$, $w = e^2 \tilde{\rho} N^{\epsilon/2}$. We showed that if perturbation theory $u \Lambda^{-\epsilon} \ll 1$ is valid then term by term entanglements are irrelevant to the physics of long chains $L \gg 1$ providing the overlap constraints

$$m^2 / \tilde{\rho} \tilde{w} \ll L^{3/2} \tag{5.3}$$

and

$$m^2 / \tilde{\rho} \tilde{w} \ll L^{d/2}, \tag{5.4}$$

arising from the modifications to Δ and \bar{G} induced by entanglements, are respected. Asymptotically for $L \gg 1$, however, the estimate (5.4) will fail through the growth of the dimensionless couplings $z = u N^{\epsilon/2}$, $w = e^2 \tilde{\rho} N^{\epsilon/2}$ ($d < 4$) characterising the perturbative domain $z \ll 1$, so we shall seek here an alternative representation for \bar{G} valid in the swelled or excluded volume limit $z = u N^{\epsilon/2} \gg 1$ of dilute physics.

To describe the asymptotic long-chain $L \gg 1$ properties of the entangled excluded volume statistics, an appropriate technique is the renormalisation group in $d = 4 - \epsilon$ dimensions as formulated by BLZ. We find that in terms of the dimensionless parameters $\bar{u} = u \Lambda^{-\epsilon}$, $\bar{x} = e^2 \tilde{\rho} \Lambda^{-\epsilon}$ the scaling function $M \equiv N^{\epsilon/2} \bar{G}$ satisfies the following renormalisation group equation in the critical domain $q/\Lambda \ll 1$, $L \gg 1$ of dilute physics $u \tilde{\rho} N \ll 1$:

$$\left[\Lambda \frac{\partial}{\partial \Lambda} + W(\bar{u}, \bar{x}) \frac{\partial}{\partial \bar{u}} + P(\bar{u}, \bar{x}) \frac{\partial}{\partial \bar{x}} + Q(\bar{u}, \bar{x}) N \frac{\partial}{\partial N} - 2Q(\bar{x}, \bar{u}) \right] M = \Delta M. \tag{5.5}$$

Here the function ΔM is smaller than M by terms of order L^{-1} , q/Λ up to powers of $\ln L$, $\ln(q/\Lambda)$ in the ϵ expansion. Perturbatively a simple analysis shows that the functions P, Q, W are of the form

$$W = -\epsilon \bar{u} + B \bar{u}^2 - C \bar{x} \bar{u} + D \bar{x}^2 + O(\bar{u}^3, \bar{x}^3), \tag{5.6}$$

$$P = -\epsilon \bar{x} - C \bar{x}^2 + O(\bar{x}^3, u \bar{x}^2), \tag{5.7}$$

$$Q = -A \bar{u} - C \bar{x} + O(\bar{x}^2, \bar{u}^2), \tag{5.8}$$

where A, B, C, D are positive real numbers and we take $\bar{u}, \bar{x} = O(\epsilon)$.

To solve the system (5.5), (5.6), (5.7), (5.8) for the scaling function $M = N^{-\epsilon/2} G$ we shall follow BLZ and observe that ΔM does not modify the behaviour of the dominant contribution $\bar{M} = N^{-\epsilon/2} \bar{G}$ (cf (4.6) *et seq.*), so in order to determine the structure of the leading term \bar{G} we may set $\Delta M = 0$ and integrate the simpler homogeneous equation ((5.5), $\Delta M = 0$). Writing $\Lambda(\tau) = \Lambda e^\tau$ and introducing functions $\bar{u}(\tau)$, $\bar{x}(\tau)$ and $N(\tau)$ such that

$$d\bar{u}(\tau)/d\tau = W(\bar{u}(\tau), \bar{x}(\tau)), \quad \bar{u}(1) = \bar{u}, \tag{5.9}$$

$$d\bar{x}(\tau)/d\tau = P(\bar{u}(\tau), \bar{x}(\tau)), \quad \bar{x}(1) = \bar{x}, \tag{5.10}$$

$$d \ln N(\tau)/d\tau = Q(\bar{u}(\tau), \bar{x}(\tau)), \quad N(1) = N, \tag{5.11}$$

then the reader will observe directly that the homogeneous equation ((5.5), $\Delta M = 0$) may be formally integrated to give the relation

$$\bar{G}(q^2, \bar{x}, \bar{u}, N, \Lambda) = (N/N(\tau))^{d/2} \bar{G}(q^2, \bar{x}(\tau), \bar{u}(\tau), N(\tau), N(\tau)) \tag{5.12}$$

through which we may analyse the scaling properties of \bar{G} and whence $\bar{C}_m(q, N)$, (5.2).

Typically for $L \gg 1$ we might expect \bar{G} to exhibit universal scaling of the canonical form

$$\bar{G} = L^{d(2\bar{\nu}-1)/2} F(q^2 L^{2\bar{\nu}}/\Lambda^2) \tag{5.13}$$

through the appearance (see BLZ) of an infrared stable fixed point \bar{u}^* , \bar{x}^* of the renormalisation group equations (5.9)–(5.12), defined as usual as solutions of the equations

$$W(\bar{u}^*, \bar{x}^*) = P(\bar{u}^*, \bar{x}^*) = 0 \tag{5.14}$$

for which the stability matrix T is positive definite,

$$T = T(\bar{u}^*, \bar{x}^*) \equiv \begin{bmatrix} \partial P/\partial \bar{u} & \partial P/\partial \bar{x} \\ \partial Q/\partial \bar{u} & \partial Q/\partial \bar{x} \end{bmatrix}_{\bar{u}=\bar{u}^*, \bar{x}=\bar{x}^*}. \tag{5.15}$$

In the presence of such fixed points a direct linearisation of the trajectory equations (5.9)–(5.11) shows that as the effective cut-off Λe^τ appearing on the right-hand side of (5.12) approaches the scales $q \ll \Lambda$, $L \gg 1$ of interest through the limit $\tau \rightarrow -\infty$, the couplings $\bar{u}(\tau)$, $\bar{x}(\tau)$ tend to their fixed point values. To generate (5.13) we then simply observe that through the functional dependence $\bar{G} = \bar{G}(q^2 N, e^2 \rho N^{\epsilon/2}, u N^{\epsilon/2})$ we may employ (5.12) and the constraint $L(\tau) \equiv N(\tau) \Lambda^2 e^{2\tau} = 1$ to show that providing both $z = u N^{\epsilon/2}$, $w = e^2 \rho N^{\epsilon/2}$ are large the function \bar{G} takes the universal scaling form (5.13), where both the critical exponent $\bar{\nu} = (2 + Q(\bar{u}^*, \bar{x}^*))^{-1}$ and the function $F(x)$ are independent of the physical couplings \bar{u} , $\bar{x} \sim e^2$. For $L \gg 1$ we therefore might reasonably expect the integration over the charge e^2 to trivialise in the sense that asymptotically \bar{G} is independent of e^2 (see (5.13)), so entanglements modify only the uninteresting amplitude A , (4.3), as we found for $d > 4$ dimensions.

Regrettably *no such fixed point exists* within the above framework, for if $P = 0$ equations (5.6), (5.7) imply $W > 0$ unless $\bar{x} = 0$ and the origin $\bar{x} = 0$, $\bar{u} = 0$ is always unstable, so that the required limit $\tau \rightarrow \infty$ is not easily characterised. Generally we see that the unstable runaway $\bar{u}(\tau)$, $\bar{x}(\tau)$ as $\tau \rightarrow -\infty$ exhibited by the lowest-order renormalisation group equations $P \sim -\epsilon \bar{u}$, $\sim \epsilon \bar{x}$ ($d < 4$) is uncontrolled, so it is tempting to suggest that this instability heralds the dominance of some perturbative fixed point or, even more catastrophically, a complete breakdown of universality; however the resolution of our problem is less dramatic.

Returning to our discussion of the perturbative domain $u\Lambda^{-\epsilon} \ll 1$ and the failure of our second estimate (5.4) through the growth of the dimensionless couplings $z = uN^{\epsilon/2}$, $w = e^2\tilde{\rho}N^{\epsilon/2}$, we observe that at least for d near four where the saddle point evaluation of the charge integral is valid (m finite) we may within the gaussian approximation of § 4 partition (5.2) in the form

$$\begin{aligned} \bar{C}_m(q, N) = 2N^{-d/2} \frac{\exp[-L\Delta(0, \bar{u})]}{(L\tilde{\rho}\Lambda^{-\epsilon})^{1/2}} & \left[\int_0^{L^{(d-2)/4}} dy \exp[-y^2(\bar{\omega}\Lambda^\epsilon)] \cos\left[\left(\frac{m^2}{L\tilde{\rho}\Lambda^{-\epsilon}}\right)^{1/2} y\bar{G}\right] \right. \\ & \left. + \int_{L^{(d-2)/4}}^\infty dy \exp[-y^2(\bar{\omega}\Lambda^\epsilon)] \cos\left[\left(\frac{m^2}{L\tilde{\rho}\Lambda^{-\epsilon}}\right)^{1/2} y\bar{G}\right] \right] \end{aligned} \quad (5.16)$$

where only in the second term do we probe the asymptotic domain $w \equiv e^2\tilde{\rho}N^{\epsilon/2} \gg 1$, all z , which exhibits the renormalisation group instability. Here $y \equiv Le^2\tilde{\rho}\Lambda^{-\epsilon}$ and $\bar{\omega}\Lambda^\epsilon$ (dimensionless) characterises the short-range or non-universal correlations in our approximation scheme as before (cf (4.17)). We see therefore that for $w \gg 1$ where the renormalisation group instability is important the non-universal prefactor is exponentially small, $\exp(-L^{(d-2)/4})$ ($\bar{\omega}\Lambda^\epsilon \sim 1$, independent of L , $d > 2$), so in the absence of more detailed information about the competition between the factor $\exp(-L\Delta)$ and the qualitatively divergent perturbation \bar{G} expansion of \bar{G} for $w \gg 1$ (all z) we may reasonably neglect this term and take

$$C_m(q, N) = 2N^{-d/2} \frac{\exp[-L\Delta(0, \bar{u})]}{(L\tilde{\rho}\Lambda^{-\epsilon})^{1/2}} \int_0^{L^{(d-2)/4}} dy \exp[-y^2(\bar{\omega}\Lambda^\epsilon)] \cos\left[\left(\frac{m^2}{L\tilde{\rho}\Lambda^{-\epsilon}}\right)^{1/2} y\bar{G}\right] \quad (5.17)$$

up to exponentially small corrections for $L \gg 1$. The evaluation of this remaining integral is still non-trivial, for as L increases we prove in turn both the perturbative $z \ll 1$ and swelled chain $z \gg 1$ limits of the excluded volume statistics. To determine the scaling form for \bar{G} appropriate to the domain of the unstable point $\bar{x}^* = 0$ (or $w \ll 1$) proved by (5.17) but otherwise valid for all values of $z = \bar{u}L^{\epsilon/2}$, we may employ the truncated renormalisation group equations (5.9)–(5.11) for which $C = D = 0$. Integrating these trajectory equations directly, we obtain the expressions

$$\left(\frac{\bar{u}^* - \bar{u}(\tau)}{\bar{u}^* - \bar{u}}\right)^{-\epsilon\nu/\omega} \frac{\bar{u}(\tau)}{\bar{u}} = e^{-\epsilon\tau}, \quad (5.18)$$

$$N(\tau) = N \left(\frac{\bar{u}^* - \bar{u}(\tau)}{\bar{u}^* - \bar{u}}\right)^{(1-2\nu)/\omega}, \quad (5.19)$$

$$x(\tau) = \bar{x} e^{-\epsilon\tau} \rightarrow \infty \quad \text{as } \tau \rightarrow -\infty \quad (5.20)$$

and

$$\bar{G}(q^2, \bar{u}, \bar{x}, \bar{N}, \Lambda) = (N/N(\tau))^{d/2} \bar{G}(q^2, \bar{u}(\tau), \bar{x}(\tau)N(\tau), \Lambda e^\tau) \quad (5.21)$$

where $\nu = (2 + Q(\bar{u}^*, 0)^{-1})$, $\omega = \nu W'(\bar{u}^*, 0)$ are respectively the usual correlation length and crossover exponents associated with the Wilson–Fisher fixed point $\bar{u}^* = (\epsilon/B) + O(\epsilon^2)$.

To derive the desired scaling relations for \bar{G} we now employ the freedom in the rescaling parameter τ by choosing $L(\tau) \equiv N(\tau)\Lambda^2(\tau) = 1$, so that the right-hand side of the fundamental equation (5.12) of (5.21) describes the physics of short $L \sim 1$ polymers

and therefore may realistically be evaluated perturbatively in the dimensionless couplings $z(\tau) \equiv \bar{u}(\tau)L(\tau)^{\epsilon/2}$, $w(\tau) \equiv \bar{x}(\tau)L(\tau)^{\epsilon/2}$ ($L(\tau) \sim 1$). Specialising to $\mathbf{q} = \mathbf{0}$ for simplicity and recalling the perturbative structure (4.11) exhibited by the dilute solution, we find using (5.21) that

$$\bar{G}(\bar{u}, \bar{x}, N, \Lambda) = \tilde{A} \left(\frac{N}{N(\tau)} \right)^{d/2} (1 + \tilde{B}\bar{u}(\tau)) \left(1 + \sum_{k=1}^{\infty} \tilde{C}_k x(\tau)^k \right) (1 + O[(\epsilon)^2]) \tag{5.22}$$

where \tilde{A} , \tilde{B} , \tilde{C}_k are numerical coefficients. Eliminating τ from (5.22) through (5.18)–(5.20) and the constraint $L(\tau) = 1$, we may then express \bar{G} in the parametric form

$$\bar{G}(\bar{u}, \bar{x}, N, \Lambda) = \tilde{A} \left[\left(\frac{\bar{u}}{p(z)} \right)^{2/\epsilon} L \right]^{d(2\nu-1)/2} (1 + \tilde{B}p) \left\{ 1 + \sum_{k=1}^{\infty} \tilde{C}_k \left[\bar{x} L^{\epsilon\nu} \left(\frac{p}{\bar{u}} \right)^{1-2\nu} \right] \right\} \tag{5.23}$$

where the parameter $p(z) \equiv \bar{u}(\tau)$ describes the dependence of \bar{G} on $\bar{z} \sim z$ through the equation

$$(1 - p/\bar{u}^*)^{-\epsilon/2\omega} p = (1 - \bar{u}/\bar{u}^*)^{-\epsilon/2\omega} \bar{u} L^{\epsilon/2} \equiv \bar{z} \sim z = \bar{u} L^{\epsilon/2}. \tag{5.24}$$

In particular for $z \gg 1$ we may solve (5.24) in the form $p = \bar{u}^*(1 + O(\bar{z}^{-2\omega/\epsilon}))$, so that asymptotically p assumes its fixed point value and we find that \bar{G} exhibits universal scaling behaviour with critical exponents ν, ω ,

$$\bar{G}(\bar{u}, \bar{x}, N, \Lambda) = \tilde{A} \left[\left(\frac{\bar{u}}{\bar{u}^*} \right)^{d(2\nu-1)/2} (1 + \tilde{B}\bar{u}^*) \left[1 + \sum_{k=1}^{\infty} \tilde{C}_k \bar{x} L^{\epsilon\nu} \left(\frac{\bar{u}^*}{\bar{u}} \right)^{1-2\nu} \right] \right] \tag{5.25}$$

where the \bar{x} dependence, although naively non-perturbative for $L \gg 1$, is suppressed through the charge integration by the non-universal prefactor $\exp(-L\Delta)$ evident in (5.17) to give effectively $\bar{x} = O(1/L)$. Naturally solving (5.24) for $z \ll 1$ we find $p \sim z$, and the perturbative results of § 4 appropriate to the random flight regime $z \ll 1$ are reproduced.

To achieve the scaling form for C_m ((5.2), $\mathbf{q} = \mathbf{0}$) of particular interest, we may now introduce the representation for $\bar{G}(z)$, (5.22), (5.24), into (5.17) to give up to exponentially small corrections the expression

$$\frac{C_m(z)}{C(z)} = \frac{1}{(\pi L)^{1/2} \tilde{\rho} \tilde{\omega}} \exp\left(-\frac{m^2}{L\tilde{\rho}\tilde{\omega}}\right) \left[1 + \frac{\tilde{C}}{L\tilde{\omega}} \left(1 - \frac{m^2}{L\tilde{\rho}\tilde{\omega}} \right) L^{\epsilon\nu} \left(\frac{p}{\bar{u}} \right)^{1-2\nu} \right] \tag{5.26}$$

where $C(z)$ is the topologically unconstrained configuration sum (say $e^2 = 0$)

$$C(z) = e^{-L\Delta(0, \bar{u})} \tilde{A} \Lambda^d \left[\left(\frac{p(z)}{\bar{u}} \right)^{2/\epsilon} L \right]^{vd} [1 + \tilde{B}p(z)], \tag{5.27}$$

and the variable $p(z)$ is to be eliminated in terms of z through (5.24). For clarity we have artificially set $\tilde{C}_k = 0, k > 1$, and ignored the modifications induced by Δ for which the estimates (4.21) *et seq.* are unchanged. We should perhaps remark that our approximation scheme is self-consistent in that $C_m(x), C(z)$ are related by the ‘integral’ sum rule

$$C(z) = \int_{-\infty}^{\infty} dm C_m(z). \tag{5.28}$$

Most generally we find therefore that entanglements do not drastically modify the

nature of the polymer statistics providing the winding number m does not violate the bounds

$$m^2 \ll L^{2-\epsilon\nu} \bar{\rho} \bar{\omega}^2 (p(z)/\bar{u})^{2\nu-1}, \quad \text{all } z, \tag{5.29}$$

and

$$m^2 \ll L^{2-1/k} \bar{\rho} \bar{\omega} (\bar{\omega}/(\bar{\omega}_k)^{1/k}), \quad k \geq 2, \tag{5.30}$$

where in the perturbative region $z \ll 1$ associated with random flight statistics (5.29) reduces to our previous result (4.24), whilst the estimate (5.30) is unchanged (see (4.22)).

To illustrate the nature of the modified bound (5.29) it is useful to consider the structure of the mean square polymer size $\langle R^2 \rangle_m$, (2.6). Observing that

$$\langle R^2 \rangle_m \equiv -\frac{1}{2d} \frac{\partial}{\partial q^2} \ln C_m(q)|_{q=0} \tag{5.31}$$

we may in direct analogy with the calculation of C_m employ (5.2), (5.21) to show that for dilute ($\rho_v = u\bar{\rho}N \ll 1$) conditions the Brereton and Shah (1981) expression (4.29), (4.30) must be replaced by the form

$$\frac{\langle R^2 \rangle_m}{\langle R^2 \rangle} = \left[1 - \frac{\bar{C}}{L\bar{\omega}} \left(1 - \frac{m^2}{L\bar{\rho}\bar{\omega}} \right) L^{\epsilon\nu} \left(\frac{p}{\bar{u}} \right)^{1-2\nu} \right] \tag{5.32}$$

where $\langle R^2 \rangle$, (2.7), is the topologically unconstrained polymer size

$$\langle R^2 \rangle = \bar{A} \left(\frac{L}{\Lambda^2} \right) \left[\left(\frac{\bar{u}}{p(z)} \right)^{2/\epsilon} L \right]^{2\nu-1} [1 + \bar{B} p(z)] \tag{5.33}$$

and $p(z)$ is to be eliminated in terms of $z = \bar{u}L^{\epsilon/2}$ by means of the relation (5.24). Here $\bar{A}, \bar{B}, \bar{C}$ are numerical coefficients and we have again for simplicity ignored the \bar{x}^k , $k > 1$, terms arising from the topological modifications to Δ, \bar{G} ; see (5.27) *et seq.*

Providing the bounds (5.29), (5.30) are respected, we thus find that entanglements are unimportant in the sense that

$$\langle R^2 \rangle_m \sim \langle R^2 \rangle \sim L^{2\nu}, \tag{5.34}$$

so we observe universal behaviour described by the usual (or linear polymer) exponents. Here $\nu = \frac{1}{2}$ for random flight statistics or $\nu \sim \frac{2}{3}$ (Flory 1969) for swelled chains.

For the physical entanglement problem ($d = 3$) we may now use (5.33) to rephrase the bounds (5.29), (5.30) in the form

$$\begin{aligned} m^2 (\langle R^2 \rangle / \langle R^2 \rangle_\theta)^2 &\ll \langle R^2 \rangle^{3/2} \bar{\rho} (\bar{\omega} \Lambda)^2 \\ m^2 &\ll \langle R^2 \rangle_\theta^{3/2} \bar{\rho} (\bar{\omega} \Lambda) (\bar{\omega} / \sqrt{\bar{\omega}_2}), \end{aligned} \quad (d = 3), \tag{5.35}$$

where $\bar{\rho} = \Lambda^2 \bar{\rho}$ is the link density of the background chain \mathcal{C}_β and $\langle R^2 \rangle_\theta$ is the mean square size of the polymer loop under random flight ($u = 0$ or θ temperature conditions $u \sim (1 - \theta/T)$)

$$\langle R^2 \rangle_\theta \equiv \bar{A} (L/\Lambda^2). \tag{5.36}$$

As before $\bar{\omega}, \bar{\omega}_k$, $k \geq 2$, summarising the effect of the short-range correlations are independent of L and therefore of little interest; we take $\bar{\omega} \Lambda, \bar{\omega}_k \Lambda^k \sim 1$.

Associated with constraints (5.35) are two critical winding numbers $m_c, \tilde{m}_c,$

$$\begin{aligned}
 m_c^2 &= \langle R^2 \rangle^{3/2} (\langle R^2 \rangle_\theta / \langle R^2 \rangle)^2 \tilde{\rho}, \\
 \tilde{m}_c^2 &= \langle R^2 \rangle_\theta^{3/2} \tilde{\rho},
 \end{aligned}
 \quad d = 3, \tag{5.37}$$

which serve to define the domain in which our analysis is valid, assuming for the present that dilute conditions are observed. Of particular importance is the ratio \tilde{m}_c/m_c which indicates whether the non-universal aspects due to Δ or the long-range correlations summarised by \tilde{G} are dominant in determining the breakdown through (5.35) of our approximation scheme and the appearance of statistics for which the nature of the topological constraints are important. We see directly from (5.37), (5.33), (5.36)

$$(\tilde{m}_c/m_c)^4 = \langle R^2 \rangle / \langle R^2 \rangle_\theta = f(\bar{z}) \geq 1 \tag{5.38}$$

where $f(\bar{z})$ is a monotonic increasing function of $\bar{z} \sim \bar{u}L^{\epsilon/2}$ of the form

$$\begin{aligned}
 f(\bar{z}) &= \left[\left(\frac{\bar{u}}{p(\bar{z})} \right)^{2/\epsilon} L \right]^{2\nu-1} (1 + \tilde{B}p(\bar{z})) \\
 p(\bar{z}) &= \begin{cases} 1 + O(\bar{z}), & \bar{z} \ll 1, \\ [(\bar{u}/\bar{u}^*)^{2/\epsilon} L]^{2\nu-1} (1 + O(\bar{z}^{-2\omega/\epsilon})), & \bar{z} \gg 1. \end{cases}
 \end{aligned}$$

Broadly then, for asymptotically long chains ($\bar{z} \gg 1$) $m_c \ll \tilde{m}_c$, so apparently the failure of our approximation scheme for winding numbers $\geq m_c$ does not indicate a breakdown of universality, but rather the inability of the renormalisation group in $d = 4 - \epsilon$ dimensions to construct the dominant singularities of \tilde{G} . In contrast, for the random flight region $z \ll 1$ the implication is somewhat different, for we observe $\tilde{m}_c/m_c \sim 1$ so that naively the artificial division between short-range and long-range correlations employed above can be misrepresentative. Apparently here the short-range details described through Δ are equally important in describing the long-range properties of the chains for winding numbers $m \geq m_c \sim \tilde{m}_c$, so we must critically review the relevance of the continuous gaussian model used, (3.1). Of course with this point of view it is therefore not surprising that the ϵ expansion, in seeking to systematically modify the random flight expressions, fails to construct the expected universal scaling forms for large winding numbers. Indeed for $\epsilon \ll 1$ we see $\tilde{m}_c/m_c(\epsilon) \ll 1$ (cf (4.22), (4.26)), in direct contrast to the result $\tilde{m}_c/m_c \geq 1$ (cf (5.38)) in $d = 3$ dimensions.

Focusing attention finally on the semi-dilute regime where the polymers strongly overlap, $u\tilde{\rho}N \gg 1$, we recall that order by order in perturbation theory topological aspects are irrelevant if

$$m^2 \ll L^{3/2} \tilde{\rho}, \quad d = 3, \quad (\text{cf (4.10), (4.23), (4.24), (4.26)}).$$

If we therefore assume that the excluded volume coupling $u\Lambda^{-\epsilon}$ is unexceptional, we may rewrite this constraint in the form

$$m^2 \ll (u\tilde{\rho}N)L^{1/2}, \tag{5.39}$$

so that for $L \gg 1$ and large overlap $o_v = u\tilde{\rho}N \gg 1$, entanglements are entirely negligible, in the sense that even deep in the critical semi-dilute regime where the length scale $(u\tilde{\rho})^{-1/2}$ grows without limit, it is difficult to see how this simple estimate might fail. Throughout the semi-dilute domain we would therefore expect that topological entanglements will not modify the large-scale or universal behaviour of the polymer system.

6. Discussion

Within the framework afforded by the partial classification of the topological aspects by the Gauss invariant $I_{\alpha\beta}$, (2.1), we have shown that the universal characteristics of a system of two long polymer loops are insensitive to the presence of topological entanglements providing the overlap constraints

$$m^2 \langle (R^2) / (R^2)_\theta \rangle^2 \ll \langle R^2 \rangle^{3/2} \bar{\rho} (\bar{\omega} \Lambda)^2, \tag{6.1}$$

$$m^2 \ll \langle R^2 \rangle_\theta^{3/2} \bar{\rho} (\bar{\omega} \Lambda) (\bar{\omega} / \sqrt{\bar{\omega}_2}), \tag{6.2}$$

are not violated. Here $\bar{\rho}$ is the link (or monomer) density of the background polymer \mathcal{C}_β and $\langle R^2 \rangle_\theta, \langle R^2 \rangle$ are the mean square sizes of the smaller polymer loop \mathcal{C}_α under random flight $u = 0$ or swelled chain $u \neq 0$ conditions ($L \gg 1$) respectively. The functions $\bar{\omega}, \bar{\omega}_2$ independent of L summarising the effect of the short-range or non-universal correlations are directly related to the ‘stiffness’ of the polymer chains at scales $\sim \Lambda^{-1}$ where Λ is representative of the inverse monomer spacing.

A relation of particular interest which follows directly from the applicability of the gaussian approximation for long chains $L \gg 1$ and winding numbers m bounded by the constraints (6.1), (6.2) describes the fractional reduction in the configuration sum

$$\frac{C_m}{C} = \frac{1}{(\pi L)^{1/2} \bar{\rho} \bar{\omega}} \exp\left(-\frac{m^2}{L \bar{\rho} \bar{\omega}}\right). \tag{6.3}$$

Valid typically for $m^2 \ll L^{2-\nu} \bar{\rho}$, $\nu \sim \frac{1}{2}$ or $\frac{3}{5}$, the relation (6.3) can represent a massive reduction in the ratio C_m/C for $L \gg 1$. This result is of special value, for we may argue (cf (4.29) *et seq.*) that if the loops were originally formed together in a melt it is highly probable that showing that (6.1), (6.2) will generally be valid ($L \gg 1$) and universality will be observed. For example, studying the expansion factor $\langle R^2 \rangle_m / \langle R^2 \rangle$, we find that entanglements lead to physical expansion

$$\langle R^2 \rangle_m / \langle R^2 \rangle = 1 + C^* (m^2 / \tilde{m}_c), \quad C^* > 0, \tag{6.4}$$

in agreement with the work of Brereton and Shah (1981) on the random flight limit where the critical winding numbers are ‘degenerate’, $m_c \sim \tilde{m}_c \sim m_c^*$. For long chains formed in the melt we would thus expect to observe universality in the form

$$\langle R^2 \rangle_m \sim \langle R^2 \rangle. \tag{6.5}$$

Of course if the loops were formed separately the invariant $I_{\alpha\beta}$ must take the value $m = 0$ and the constraints (6.1), (6.2) become trivial.

Trivialising for $m = 0$ and semi-dilute systems $u\bar{\rho}N \gg 1$, our constraints reflect the difficulty of forming complex knots $m \gg 0$ without affecting the large-scale properties of the system when the overlap between the polymers is small and the chains are inflexible at scales $\sim \Lambda^{-1}$. Associated with the constraints (6.1), (6.2) are two critical winding numbers

$$m_c^2 \equiv \langle R^2 \rangle^{3/2} \langle (R^2)_\theta / \langle R^2 \rangle \rangle^2 \bar{\rho}, \tag{6.6}$$

$$\tilde{m}_c^2 \equiv \langle R^2 \rangle_\theta^{3/2} \bar{\rho}, \tag{6.7}$$

which satisfy a relation of the form

$$(\tilde{m}_c / m_c)^4 = \langle R^2 \rangle / \langle R^2 \rangle_\theta = f(\bar{z}) \geq 1 \tag{6.8}$$

where $f(\bar{z})$ is a monotonically increasing function of $\bar{z} \sim uL^{1/2}$ in the dilute domain

$u\tilde{\rho}N \ll 1$ and is effectively constant for semi-dilute physics. Typically for asymptotically long chains $L \gg 1$ and dilute conditions $u\tilde{\rho}N \ll 1$ we can estimate $(\tilde{m}_c/m_c)^4 \sim L^{2\nu-1} \gg 1$ where $\nu \sim \frac{3}{2}$ is the Flory exponent associated with the mean square size of a swelled ($\bar{z} \gg 1$) polymer loop $\langle R^2 \rangle \sim L^{2\nu}$, so the constraint (6.1) is of dominant importance. To interpret (6.1) it is useful to introduce the concept of an equivalent flexible chain familiar in polymer physics. As usual, defining an effective monomer spacing Λ^{*-1} such that

$$\langle R^2 \rangle \equiv L\Lambda^{*-2} \sim L^{2\nu}, \quad (6.9)$$

we see that the ratio

$$\langle R^2 \rangle / \langle R^2 \rangle_\theta = (\Lambda/\Lambda^*)^2$$

implies through (6.1), (6.6) that as the effective link size Λ^{*-1} increases at fixed overlap $\bar{\rho}_v = \langle R^2 \rangle^{3/2} \bar{\rho}$ the critical winding number m_c is reduced as one might expect.

The ratio \tilde{m}_c/m_c is of particular importance for it indicates whether the non-universal aspects due to Δ or the long-range correlations described by \bar{G} are dominant in determining the breakdown of our approximation scheme. For swelled chains $\bar{z} \gg 1$ we have already noted above that $\tilde{m}_c/m_c \gg 1$ which would indicate that for winding numbers $m \leq m_c \ll \tilde{m}_c$ universality will be observed but whose description lies outside of the framework defined by the renormalisation group adopted in § 5. In contrast, for the random flight $\bar{z} \ll 1$ regime the implication is somewhat different, for we observe $\tilde{m}_c/m_c \sim 1$ so that the artificial division between short-range and long-range correlations employed above, (4.6), (4.7), can be misrepresentative. Apparently here the short-range correlations described through Δ are equally important in describing the long-range properties of the chains for $m \leq m_c \sim \tilde{m}_c$, so to describe the physics we must first critically review the relevance of the phenomenological gaussian model employed.

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