Journal of Mathematical Chemistry 8(1991)255–267

ON THE BEHAVIOUR OF COLLAPSING LINEAR AND BRANCHED POLYMERS

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Abstract

In this paper, we have reviewed the behaviour of a variety of models of collapse transitions in linear and branched polymers. Both rigorous and numerical results are presented. Some of the work described is still in progress, so the results are incomplete. Nevertheless, the overall picture demonstrates how the association of an attractive fugacity between nearby monomers can give rise to a collapse transition in a wide variety of models. These then qualitatively describe the transition undergone by collapsing linear and branched polymers.

1. Introduction

We discuss a number of recent studies of models of collapse in both linear and branched polymers. We present both rigorous results in the form of existence proofs and bounds on thermodynamic quantities, numerical results used to locate the positions and identify the nature of the collapse transition, and exact results for a simplified model that nevertheless retains the essential features of a more complicated model. We also consider the effect of particular geometrical constraints on the linear polymer system.

A linear polymer molecule in dilute solution in a good solvent can be modelled by a self-avoiding walk (saw) on a regular lattice. By suitably weighting near-neighbour interactions with an appropriate fugacity, the (infinite) walk is believed to undergo a transition which adequately models the transition in real polymers brought about by the dominance of attractive forces at low temperatures or a poor solvent. When the fugacity is neutral, or favours near-neighbour repulsion, the walk is in a swollen phase, corresponding to the behaviour in a good solvent. As the fugacity is weighted towards attraction, the walk adopts a more compact configuration. Between these two regimes is an intermediate regime, corresponding to a tricritical point. This model, based on weighting saws has been studied theoretically by many authors [1–7]. A related model in which the collapse transition in suitably weighted self-avoiding polygons has been studied is reported in [8].

Just as saws model linear polymers, so do lattice animals model the configurational properties of randomly branched polymers in dilute solution in a good solvent. As the solvent quality deteriorates, branched polymers are expected to become more compact and a collapse transition analogous to that discussed above is expected to occur. In these models, one associates a fugacity with every cycle, so that the energy is proportional to the cyclomatic index. Making this energy suitably attractive is then expected to lead to a collapse phenomenon, since animals with many cycles will be more compact than those with few cycles. This model has also attracted considerable attention [9 – 14]. For a directed animal problem, it can be proved that there is a collapse transition [15].

In subsequent sections, we describe our work on several models and report the results obtained to date. In some cases this is ongoing work, and additional results are expected to be forthcoming and will be published subsequently.

2. Walks crossing a square

In this problem, we consider self-avoiding walks on the square lattice which are confined to lie in or on the boundary of a square with vertices at $(0, 0)$, $(0, L)$, $(L, 0)$, and (L, L). We first consider the number of such walks that begin at the origin and end at the vertex (L, L), especially in the large L limit. At fixed L, we also associate a fugacity with the number of steps in the walk and ask how the system behaves as a function of this fugacity.

Both problems have been investigated both analytically and by series analysis. We denote by $c_n(L)$ the number of saws with n steps confined to lie in the $L \times L$ square and which start at the origin and finish at (L, L). Denote by $c(L)$ the sum $\sum_n c_n(L)$. By considering the maximum and minimum value of n for fixed L, we can prove that

$$\lim_{L \to \infty} \sup L^{-2} \log c(L) \leq \log \mu, \tag{2.1}$$

where μ is the growth constant for saws on the square lattice. By partially covering the $L \times L$ square with smaller squares of side $M + 2$, it is possible to prove by concatenation arguments that the limit

$$\lim_{L \to \infty} L^{-2} \log c(L) = \log \lambda \tag{2.2}$$

exists. We obtain a lower bound on $c(L)$ by observing that, for L even, walks with the maximum number of steps are Hamiltonian walks. A lower bound for the number of such Hamiltonian walks is obtained by a construction similar to that used by Gujrati [26]. The square is effectively covered by disjoint rectangles of size $(1 \times m)$. We consider all possible ways of joining these rectangles so as to form Hamiltonian polygons, and this gives the bound

$$\log \mu_H \geq \log m/[2(m+1)]. \tag{2.3}$$

This bound is most effective when $m = 4$, when it is equal to 0.1386... . Finally, to convert the polygon to a walk crossing the square we remove and insert appropriate edges. This does not change the bound, so that we finally have:

$$\lim_{L \to \infty} L^{-2} \log c(L) \geq 0.1386\ldots . \tag{2.4}$$

That is to say, we have proved that the number of distinct self-avoiding paths grows exponentially with the *area* of the lattice, rather than exponentially with its linear dimension. Similarly, the mean number of steps in such a path $\langle n \rangle = \sum_n n c_n(L)/\sum_n c_n(L)$ must be of order L^2, as can be seen by the following argument. Firstly, observe that $c_n(L) \leq \mu^{n+o(n)}$. If $n = o(L^2)$, then $c_n(L) \leq \mu^{o(L)}$ so that all except exponentially few walks have of order L^2 steps. Hence, the mean number of steps must also be of order L^2.

Numerically, we have derived exact values of $c_n(L)$ for $L \leq 6$. Summing over n, we obtain $c(L)$ values which, when extrapolated, permit us to estimate $\lambda = 1.75 \pm 0.01$.

For the second aspect of this problem, we introduce a step fugacity x, and consider the generating function

$$C_L(x) = \sum_n c_n(L) x^n. \tag{2.5}$$

By a refinement of the proof discussed above, we can prove the existence of the limit

$$\lim_{L \to \infty} L^{-2} \log C_L(x) = \log \lambda(x). \tag{2.6}$$

To estimate $\lambda(x)$, we first note that for $x \leq 1$ an upper bound is obtainable by observing that every walk which crosses the square is "doubly unfolded" (that is, the end-points are the "top" and "bottom" points). Since such walks can be concatenated to give a supermultiplicative inequality, we have $c_n(L) \leq \mu^n$ and hence that

$$C_L(x) \leq \sum \mu^n x^n = (\mu x)^{2L}(1 - (\mu x)^{n_{\max} - 2L + 1})/(1 - \mu). \tag{2.7}$$

If $x > 1/\mu$, then (2.7) implies that $\log \lambda(x) \leq \log \mu + \log x$. If $x < 1/\mu$, we have that $\log \lambda(x) \leq 0$, and combined with the bound $C_L(x) \geq c_{2L}(L) x^{2L}$, this implies that $\log \lambda(x) = 0$ for all $x \leq 1/\mu$. Hence, $\log \lambda(x)$ is non-analytic. To determine the point of non-analyticity, we note that $C_L(x) \geq c_{n_{\max}}(L) x^{n_{\max}}$, and so $\log \lambda(x) \geq \log \mu_H + \log x$, where μ_H is defined above. Thus, there must be a singular point x^* in the range $1/\mu \leq x^* \leq 1/\mu_H$. To investigate this numerically, we have studied the mean number as a function of fugacity. We define this quantity by

$$\langle n(x,L)\rangle = \sum_n n c_n(L)x^n / \sum_n c_n(L)x^n \tag{2.8}$$

and expect that $\langle n(x, L)\rangle = A(x)L^2[1 + o(1)]$. We have estimated $A(x)$ for a range of values of x, and found that it vanishes between $x = 0.3$ and $x = 0.4$. From the above bounds on x^*, we have that $0.37905 \le x^* \le 0.1386$. It is thus tempting to suggest that $x^* = 1/\mu$ exactly, although we have been unable to prove (or disprove) this suggestion. Our numerical data are insufficient to estimate the critical exponents associated with the transition. Further details of the calculation will be published elsewhere [16].

3. Collapsing branched polymers

Here, we consider a lattice model of a branched polymer in dilute solution in which the polymer is modelled both by weakly embeddable lattice animals and by trees. The models are motivated by the idea that randomly branched polymers can clearly be modelled by animals, while work by Lubensky and Isaacson [9] suggests that cycles are relatively unimportant, so this suggests the simpler model of a lattice tree. Details of the models and this work will appear [17, 18], and here we present a summary of the main results.

A *site*-animal is a connected section graph of the underlying lattice, and we denote the number of these as A_n (B_n), where n refers to the number of vertices (edges). A *bond*-animal is a connected subgraph of the underlying lattice, and we denote the number of these as a_n (b_n), where again n refers to the number of vertices (edges). Trees are acyclic animals, and we denote by T_n and t_n the number of site and bond trees, respectively, with n vertices (and hence $n - 1$ edges). In each of the six cases above, concatenation arguments enable us to establish the existence of the limit

$$\lim_{n\to\infty} n^{-1}\log X_n = \log \lambda_x\,, \tag{3.1}$$

where X_n refers to any of the six quantities defined above. The corresponding *growth constants* λ_x are known to be finite and non-zero. In order to study the collapse transition, we associate an energy with contacts, where a contact is defined to be two adjacent vertices not joined by an edge. This definition clearly precludes site animals and site trees. If $X_n(u)$ denotes the number of bond *beasts* (animals with n vertices, animals with n edges, or bond trees), the corresponding partition function and free energy are given by

$$Z_n(\beta;X) = \sum_u X_n(u)\exp(\beta u) \tag{3.2}$$

and

$$G(\beta;X) = \lim_{n\to\infty} n^{-1}\log Z_n(\beta;X), \tag{3.3}$$

respectively. Alternative models arise if we weight not by an energy associated with the number of contacts, but with the cyclomatic index. In that way, (3.2) and (3.3) have "u" replaced by "c", where c is the cyclomatic index. Since trees have cyclomatic index zero, this model is restricted to animals (now incuding site animals). We denote the partition function and free energy as above, with superscript "0" to indicate this model. Thus, we have three models in which we weight by contacts and four models in which we weight by cyclomatic index.

For all seven models, we have proved that the free energy exists, that it is convex, monotone and continuous. Useful bounds on the free energy have also been derived.

Numerical studies have established the β dependence of the free energy, based on exact enumeration data. In particular, Gaunt and Flesia [14] have examined the temperature dependence of the heat capacity for various values of n, and have estimated the location of the phase transition by extrapolating the locations of the maxima in these curves.

4. A directed walk model

In this work, we considered a variant of the interacting saw model which is exactly solvable. On the square lattice, we consider saws in which no step in the negative x direction are allowed. In the non-interacting case, this is a straightforward model to solve. The generating function for n-step walks is singular at $(1+\sqrt{2})^{-1}$, with a simple pole singularity at that point. However, if we consider the number of n-step walks with m near-neighbour contacts $c_n(m)$, the problem is more difficult, but can still be exactly solved [19 – 22]. To simplify certain calculations, we impose the additional technical condition that the first step of the walk is in the positive x-direction.

A number of rigorous results can be obtained by concatenation arguments. Consider the generating function

$$Z_n(x) = \sum_m c_n(m) x^n \tag{4.1}$$

and the corresponding quantity

$$\kappa(x) \equiv \lim_{n\to\infty} n^{-1} \log Z_n(x), \tag{4.2}$$

then, as noted above, $\kappa(1) = \log(1+\sqrt{2})$. Concatenation arguments, plus the observation that $Z_n(x)^{1/n}$ is bounded above for $x < \infty$, are sufficient to establish the existence of the limit (4.2), and its finiteness for $x < \infty$. Further, the functional inequality derived by concatenation implies that $\kappa(x)$ is log-convex and hence continuous. From the observation of monotonicity of $\kappa(x)$ in both the regimes $x < 1$ and $x > 1$, plus the fact that $\max(m) = n + o(n)$, we prove that

$$\lim_{x \to 0^+} \kappa(x)/\log x = 0, \qquad \lim_{x \to +\infty} \kappa(x)/\log x = 1. \tag{4.3}$$

We shall find it useful to define the two-variable generating function

$$G(x,y) = \sum_{n,m} c_n(m) x^m y^n. \tag{4.4}$$

At fixed x, $G(x, y)$ converges for $y < \exp(-\kappa(x))$, which defines the phase boundary $y = y(x)$ in the (x, y) plane. From (4.3) it follows that, for large x, $y \sim 1/x$.

We now use the technique of Temperley [23] to derive an explicit expression for $G(x, y)$. Let $c_n(r, m)$ denote the number of n-step walks with m contacts and the first step in the positive x direction and then exactly r steps in the y direction (the $(r+1)$th step must be in the positive x direction). We define the generating function

$$g_r(x,y) = \sum c_n(r,m) x^m y^n \tag{4.5}$$

and then write down recurrences for the g_r for $r \geq 0$. By eliminating terms between the g_r we obtain the recurrence relation

$$g_{n+1} - (1+x)y\, g_n - (1-x)x^n y^{n+2} g_n + xy^2 g_{n-1} = 0. \tag{4.6}$$

After some manipulation, we obtain

$$G = [2y g_1' - (2+y-xy)g_0']/[y^2(1+x+y+xy)g_0' - 2y g_1'], \tag{4.7}$$

where

$$g_n' = y^n + y^n \sum_{m=0}^{\infty} z^{mn}(y-z)^m z^{m(m+1)/2} \Big/ \left[\prod_{k=1}^{m} (z^k - 1)(y z^k - z) \right] \tag{4.8}$$

and $z = xy$.

The phase boundary is the locus of singular points of G closest to the x-axis for positive y. An analysis of g_0' and g_1' shows that they are analytic except on the hyperbola $z = 1$. Hence, from (4.7) we see that G is analytic except on this hyperbola and at zeros of the denominator of (4.7). The recurrence relation (4.6) can also be solved on the hyperbola $z = 1$, where

$$G(\tfrac{1}{y}, y) = y(1+\lambda)/(1-\lambda-y(1+\lambda))$$

and (4.9)

$$\lambda = \tfrac{1}{2}\{1 + y^2 - (1 - 4y + 2y^2 + y^4)^{1/2}\}.$$

We have numerically determined the zeros of (4.7) which lie below the hyperbola for $x < x^* = 9/[(17 + 3\sqrt{33})^{1/3} + (17 - 3\sqrt{33})^{1/3} - 1]^2$, while for $x \geq x^*$ the phase boundary is coincident with the hyperbola $z = 1$. The point $y^* = 1/x^*$ is also

a pole of $G(y^{-1}, y)$. Physically, the significance of the phase boundary can be seen by considering the average value of n at some point (x, y), defined by

$$\langle n(x,y)\rangle = \frac{\partial \log G(x,y)}{\partial \log y}. \tag{4.10}$$

On the phase boundary $\langle n\rangle$ is infinite, while below the phase boundary it is finite. Above the phase boundary, we enter the non-physical region corresponding to $\langle n\rangle < 0$. The point (x^*, y^*) corresponds to a collapse transition, and models the coil – ball transition undergone by real polymers. Full details of this model will be reported in [22], while a number of results are already reported in [20]. A related model in which all horizontal segments are of equal length was introduced by Zwanzig and Lauritzen [27 – 29].

5. The collapse transition in two-dimensional vesicles

In this problem, we [24] enumerated self-avoiding polygons on the square lattice by both perimeter and area. By associating a fugacity with both area and perimeter, we can induce a collapse transition along a line of critical points in the two variable fugacity plane. We consider the number $p_m^{(n)}$ of polygons on the square lattice with m edges enclosing area n. The generating function for such polygons is given by

$$P(x,y) = \sum_{m,n} p_m^{(n)} x^m y^n = \sum_m P_m(y) x^m = \sum_n A_n(x) y^n, \tag{5.1}$$

where $P_m(y)$ and $A_n(x)$ are, respectively, the generating functions for polygons of perimeter m and area n. The generating function $P(x, y)$ can be considered a grand partition function.

By considering polygons with maximum area for fixed perimeter, we show that

$$\lim_{m\to\infty} m^{-2} \log P_m(y) = \log y/16 \quad \text{for all } y \geq 1. \tag{5.2}$$

For $0 < y < 1$, $\lim_{m\to\infty} m^{-1} \log P_m(y) = \kappa(y)$ exists and is finite. We note that $\kappa(1) = \kappa$, the usual connective constant for polygons grouped by perimeter.

From the observation that

$$P_m(y_1) P_m(y_2) \geq \left[P_m\left(\sqrt{y_1 y_2}\right)\right]^2, \tag{5.3}$$

it follows that $\kappa(y)$ is log-convex.

By considering the polygons of minimum area with fixed perimeter, we obtain the bound $\kappa(y) \leq \kappa + 1/2 \log y$. Observing that these are just the number of

a class of site trees on the dual lattice, we can concatenate them and hence prove the existence of the thermodynamic limit κ_0 in that case. Hence,

$$\kappa(y) \geq (\kappa_0 + \log y)/2. \tag{5.4}$$

The two bounds together imply

$$\lim_{y \to 0^+} \kappa(y)/\log y = 1/2. \tag{5.5}$$

Now the grand partition function can be written as

$$P(x, y) = \sum_m e^{mk(y)+o(m)} x^m, \tag{5.6}$$

and for fixed x this converges for $x < e^{-\kappa(y)}$, so that $x = e^{-\kappa(y)}$ defines a phase boundary. For $x > e^{-\kappa(y)}$, the free energy is not extensive. As $y \to 0$, $x \to \infty$ as

$$x \sim y^{-1/2}. \tag{5.7}$$

Further, as $\kappa(y)$ is monotone non-decreasing, $x(y)$ is monotone non-increasing. Further, $x(y)$ is bounded below by $e^{-\kappa}$ for $y \leq 1$ and then jumps discontinuously to zero. The bounds established above enable us to write

$$e^{-\kappa_0} \geq x^2 y \geq e^{-2\kappa}. \tag{5.8}$$

Similar results have been obtained for the generating function $A_n(x)$. The limit

$$\lim_{n \to \infty} n^{-1} \log A_n(x) = \chi(x) \tag{5.9}$$

exists, and we show that $\chi(x)$ is log-convex. Additional simple arguments allow us to obtain the bounds

$$\chi(1) + 2\log x \geq \chi(x) \geq \lim_{n \to \infty} n^{-1} \log p^{(n)}_{2n+2} + 2\log x \quad \text{for } x \geq 1. \tag{5.10}$$

Hence,

$$\lim_{x \to \infty} \frac{\chi(x)}{\log x} = 2. \tag{5.11}$$

For $x \leq 1$, we prove that $\chi(x) \leq 2\kappa + 2 \log x$ when $e^{-\kappa} \leq x \leq 0$, and $\chi(x) = 0$ when $x \leq e^{-\kappa}$. Similar results have been obtained for three and higher dimensions. In d dimensions, we now treat surfaces in Z^d made up of elementary unit $(d-1)$-dimensional "hypersquares". A similar phase boundary is obtained, and the analogous result to (5.7) is

$$x \sim y^{-1/(2d-2)} \quad \text{as } y \to 0^+. \tag{5.12}$$

In all dimensions, the phase boundary $x(y)$ is monotone non-increasing and jumps discontinuously to zero at $y = 1$.

In two dimensions, we have constructed an accurate numerical phase diagram by enumerating all polygons with perimeter ≤ 44 for all areas. We analysed the series for fixed y, and determined $x(y)$ in this way. The resulting phase diagram is

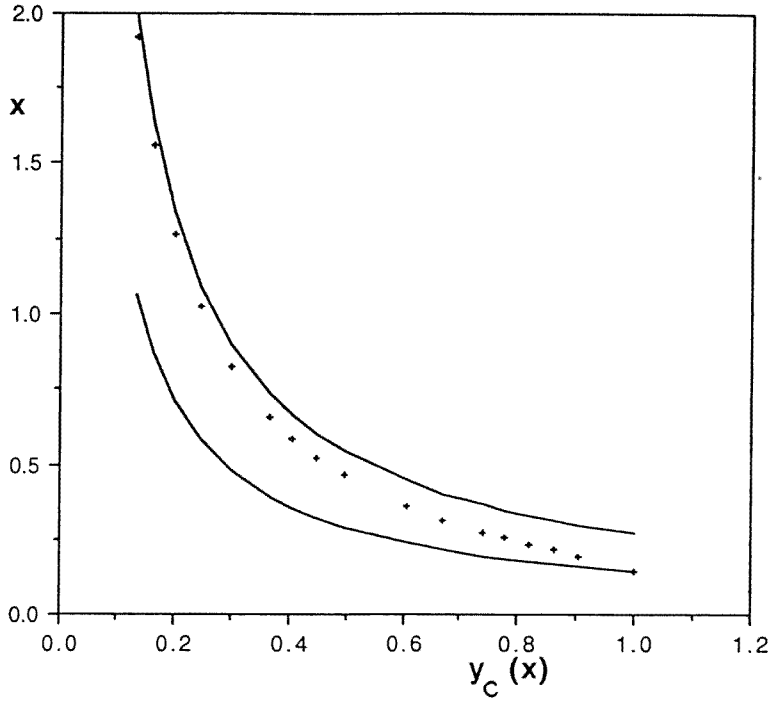

Fig. 1. Phase diagram of two-dimensional vesicles. Above the phase boundary, the free energy is no longer extensive. The solid curves are the upper and lower bounds, the broken curve is the numerically determined phase boundary.

shown in fig. 1. In the vicinity of the multicritical point, the grand partition function behaves, to leading order, as

$$P(x,y) \sim C_0 |\tilde{x}|^{2-\alpha_0} Z(\tilde{y}/|\tilde{x}|^{\phi}) + B(x,y), \tag{5.13}$$

where the scaling axes $\tilde{x}, \tilde{y}$ are given by

$$\tilde{x} = x_c - x + (y-1)/e_2, \quad \tilde{y} = 1 - y.$$

$\alpha_0 = 1/2$ is the usual polygon exponent, and $Z(z)$ is a scaling function with normalisation $Z(0) = 1$. The crossover exponent ϕ is shown to be equal to $2\nu = 3/2$, where $\nu = 3/4$ is the usual saw correlation function exponent.

6. The collapse of self-avoiding polygons

If we introduce an energy term associated with near-neighbour contacts, we would expect a collapse transition in polygons just as in walks [8]. One technical reason for studying the polygon problem is that polygons can be concatenated without (substantially) changing the total number of contacts. A second reason for studying this problem is that it allows us to investigate the influence of architecture on the collapse transition.

We consider the square lattice. Let $p_n(m)$ be the number (per lattice site) of polygons with n edges and m near-neighbour contacts. Clearly,

$$\sum_m p_n(m) = p_n\,, \tag{6.1}$$

the total number of polygons, and

$$\lim_{n\to\infty} n^{-1} \log p_n = \kappa \tag{6.2}$$

exists, which is the connective constant of the lattice. We can define the partition function

$$Z_n(\beta) = \sum_m p_n(m)\, e^{\beta m} \tag{6.3}$$

and we are interested in the properties of the corresponding limiting free energy.

Pairs of polygons can be concatenated by the addition of a "neck" [24] to give the functional inequality

$$p_n(m) = \sum_{m_1} p_n(m_1) p_{n-n_1}(m - m_1 - 2), \tag{6.4}$$

which easily gives the existence of the limit

$$\lim_{n\to\infty} n^{-1} \log Z_n(\beta) \equiv A(\beta). \tag{6.5}$$

Moreover, $A(\beta)$ is convex and hence continuous.

Clearly, $A(0) = \kappa$ and, by monotonicity, $A(\beta) \le \kappa$ for $\beta \le 0$ and $A(\beta) \ge \kappa$ for $\beta \ge 0$. For $\beta \le 0$,

$$Z_n(\beta) \ge p_n(0), \tag{6.6}$$

so

$$A(\beta) \geq \kappa_0,$$

where κ_0 is the connective constant for neighbour-avoiding walks and, for $\beta \geq 0$,

$$p_n e^{\beta m_{\max}} \geq Z_n(\beta) \geq p_n(m_{\max}) e^{\beta m_{\max}}. \tag{6.7}$$

Since $m_{\max} = n/2 + o(n)$, taking logarithms, dividing by n and letting $n \to \infty$, we have

$$\kappa + \beta/2 \geq A(\beta) \geq \max[\kappa, \kappa^* + \beta/2], \tag{6.8}$$

for $\beta \geq 0$, where $\kappa^* = \lim n^{-1} \log p_n(m_{\max})$, and hence $\lim_{\beta \to \infty} A(\beta)/\beta = 1/2$.

Although these results give useful information on the general behaviour of $A(\beta)$, they do not establish that the model shows a phase transition. To investigate this, we have derived exact values of $p_n(m)$ for $n \leq 34$, and computed $c_n(\beta) = \partial^2 A_n(\beta)/\partial \beta^2$, where

$$A_n(\beta) = n^{-1} \log Z_n(\beta),$$

for a range of values of β. In fig. 2, we show the dependence of $c_n(\beta)$ for $n = 14-34$. There is a fairly sharp peak whose position is only weakly dependent on n. By extrapolating the peak positions, we estimate that the transition occurs at $\beta_0 \approx 0.64$.

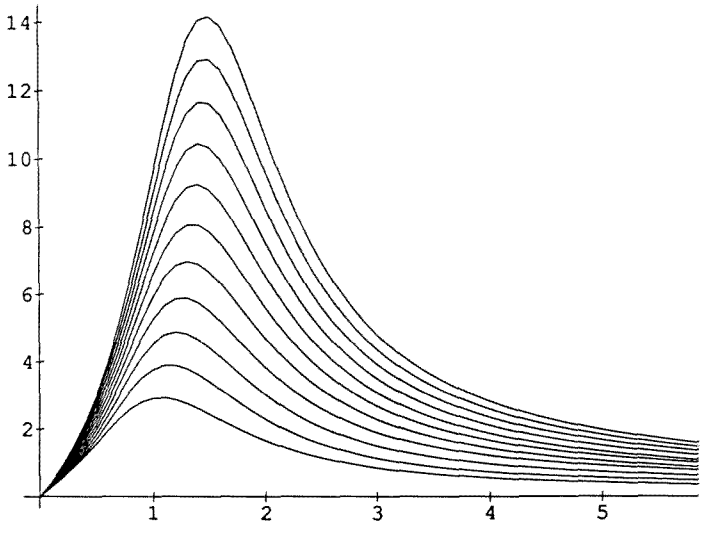

Fig. 2. Contact fluctuation as a function of attractive fugacity exp(w) for polygons of sizes 14, 16, . . ., 34.

Further numerical and Monte Carlo work is still being undertaken [25] and will be reported elsewhere.

7. Conclusion

We have described five different models of collapse transition, and shown how in each case a combination of rigorous and numerical results elucidate the nature of the collapse transition. In each case a fugacity is associated with a particular property of the model, usually the number of adjacent monomers or the area. By making this fugacity attractive or repulsive, one switches from a collapsed to an expanded state. This transition is seen in real polymeric systems in, for example, the coil–ball transition. It is hoped that the description here of a range of models will gather together a sufficiently broad range of techniques to permit subsequent models to be similarly studied.

Acknowledgements

A.J. Guttmann acknowledges the support of the Australian Research Council, and S.G. Whittington acknowledges support by NSERC of Canada. Some of this work was carried out while S.G. Whittington was visiting the Mathematics Department of the University of Melbourne, and he is grateful to the members of that department for their hospitality.

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