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Size of a 2D ring polymer topologically unentangled with a planar array of obstacles

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We readdress the statistical mechanical problem of the size of a 2D ring polymer, topologically unentangled with a planar lattice array of regularly spaced obstacles. It is commonly assumed in the literature that such a polymer adopts a randomly branched type of configuration, in order to ostensibly maximise chain entropy, while minimising obstacle entanglement. Via an innovative analytic approach, valid in the condensed polymer region, we are able to provide a greater theoretical understanding, and justification, for this presumed polymer behaviour. Our theoretically derived results could also potentially have important implications for the structure of interphase chromosomes, as well as electrophoretic ring polymer dynamics.

Keywords: ring polymer, obstacles, topological interaction.

INTRODUCTION

The intriguing role of topology, or topological constraints, in determining the properties of unentangled ring polymers continues to fascinate researchers in the field [1, 2]. Indeed, there exists a substantial literature on this topic (see e.g. [3–6] and references therein), with applications ranging from chromosomal structure [7, 8] to gel electrophoresis [9].

The purpose of this work however is to reinvestigate, or reconsider, the simpler problem of the size R of a 2D ring polymer (with degree of polymerisation N) which is topologically unentangled with a planar lattice array of regularly spaced obstacles (see Fig. 1). Previous work on this specific topic include that of [10–12].

In the work of [10], for example, a branched random lattice was invoked from the outset for the allowable polymer conformations, which naturally leads to the randomly branched polymer result: $R \sim N^{1/4}$ [13]. Whereas, in [12] a polymer area order parameter was introduced, and via the approximate summation of leading logarithmic terms, again the randomly branched polymer result was obtained (in the absence of the excluded volume interaction).

To the best of our knowledge, there appears to be a paucity of work available in the literature regarding any direct simulations of the explicitly 2D case, as considered in this work. Indeed, it has become somewhat common lore, and indeed even ‘obvious’, that an unentangled polymer in an array of obstacles adopts a randomly branched structure (possibly going over to something like a fractal or crumpled globule [5, 6] at higher densities).

Despite the previous work mentioned above, it therefore seems naturally desirable for us to restudy, and elucidate further, precisely how considerations of polymer topology can give rise to randomly branched polymer statistics. In particular, we would like to gain some additional and improved theoretical understanding on how the presence of topological constraints (via the use of a 2D linking number defined below) can lead to the typical size R of the polymer adopting a randomly branched polymer type of behaviour given by roughly $R \sim N^{1/4}$.

THEORY

We firstly proceed to provide below a concise derivation of the theory to be used. The interested reader is invited to consult [11, 12, 14–17] for more general, background, information.

The (topologically invariant and integer valued) 2D Linking Number Φ_{nm} between our polymer chain on the plane, $R^\alpha(s)$, and an isolated planar obstacle at lattice position $R_{nm}^\alpha = (na, ma)$, with lattice spacing a , is defined as [11, 12] :

$$\Phi_{nm} = \frac{1}{2\pi} \int_0^L ds \epsilon_{\alpha\beta} \frac{(R^\alpha(s) - R_{nm}^\alpha)}{(R(s) - R_{nm})^2} \frac{\partial R^\beta(s)}{\partial s} \quad (1)$$

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where s is the contour length along the chain, and $\epsilon_{\alpha\beta}$ is the 2D antisymmetric tensor (defined by $\epsilon_{12} = +1$ and $\epsilon_{21} = -1$).

For an unentangled ring polymer, we wish to introduce the constraint that the (integer valued) 2D Linking Number Φ_{nm} between our polymer chain and an isolated obstacle vanishes, as follows:

$$\begin{aligned} \prod_{nm} \delta(\Phi_{nm}) &= \prod_{nm} \frac{\sin(\pi \Phi_{nm})}{\pi \Phi_{nm}} \approx \prod_{nm} \left(1 - \frac{\pi^2}{6} \Phi_{nm}^2\right) \\ &\approx \prod_{nm} e^{-\frac{\pi^2}{6} \Phi_{nm}^2} \approx e^{-\frac{\pi^2}{6} \sum_{nm} \Phi_{nm}^2} \end{aligned} \quad (2)$$

where, in order to make analytic progress, the approximation used is to expand for values of the linking number close to zero, this being the specific regime of interest for this work.

Therefore, in order to penalise polymer conformations with non-zero linking number, we introduce the following Hamiltonian, H_C , as :

$$\begin{aligned} H_C &= \frac{\pi^2}{6} \sum_{nm} \Phi_{nm}^2 \\ &\approx \frac{\pi^2}{6a^2} \int d^2x \Phi^2(x) \end{aligned} \quad (3)$$

where in the second line, and for calculational convenience and tractability, we pass to the continuum approximation (with a short-distance cut-off a corresponding to the lattice spacing of obstacles).

The required Boltzmann factor, e^{-H_C} , (in the continuum approximation) can be simply unpacked via the introduction of a vector gauge field, $A_\alpha(x)$, as follows (for the analogous 3D case see [14–17]):

$$e^{-H_C} = \int DA_\alpha e^{-H_A + i \int_0^L ds A_\alpha(R(s)) \frac{\partial R^\alpha(s)}{\partial s}} \quad (4)$$

where, by utilising the 2D identity, $\epsilon_{\alpha\beta} \epsilon_{\mu\nu} = \delta_{\alpha\mu} \delta_{\beta\nu} - \delta_{\alpha\nu} \delta_{\beta\mu}$, the gauge field Hamiltonian, H_A , can be written as :

$$H_A = \frac{1}{2} \int d^2x \left[\frac{3a^2}{\pi^2} (\epsilon^{\alpha\beta} \partial_\alpha A_\beta)^2 + \frac{1}{\eta} (\partial_\alpha A^\alpha)^2 \right] \quad (5)$$

which includes the usual gauge fixing term, controlled typically by the parameter $\eta \rightarrow 0$ [14–17].

The familiar 2D polymer elasticity Hamiltonian [18], H_{el} , is given by :

$$H_{el} = \frac{1}{b} \int_0^L ds \delta_{\alpha\beta} \frac{\partial R^\alpha(s)}{\partial s} \frac{\partial R^\beta(s)}{\partial s} \quad (6)$$

with a step length b , while the 2D polymer excluded volume interaction is typically written [18] as (with interaction strength ν) :

$$H_{ev} = \frac{\nu}{2} \int_0^L ds \int_0^L ds' \delta^2(R(s) - R(s')) \quad (7)$$

The Boltzmann weight associated with the polymer excluded volume interaction, given by H_{ev} , can similarly be re-expressed, by introducing an auxiliary scalar field $\phi(x)$, as follows [18]:

$$e^{-H_{ev}} = \int D\phi e^{-\frac{1}{2\nu} \int d^2x \phi^2(x) + i \int_0^L ds \phi(R(s))} \quad (8)$$

We are now in a position to form the requisite Green function, $G(x, x'; L)$, as a path integral [14–17] :

$$G(x, x'; L) = \int_{R(0)=x'}^{R(L)=x} DR(s) e^{-H_{el} + i \int_0^L ds A_\alpha(R(s)) \frac{\partial R^\alpha(s)}{\partial s} + i \int_0^L ds \phi(R(s))} \quad (9)$$

which can be shown to satisfy the following governing equation [14, 17]:

$$\frac{\partial}{\partial L} G(x, x'; L) = \left(\frac{b}{4} (\partial_\alpha - iA_\alpha)^2 - i\phi \right) G(x, x'; L) \quad (10)$$

with the ring polymer case, as considered in this work, clearly corresponding to the limit: $x \rightarrow x'$.

In order to proceed, it typically proves more useful to utilise the Laplace transformed Green function (w.r.t. the polymer length L), defined as: $\tilde{G}(x, x'; \mu) = \int_0^\infty dL e^{-\mu L} G(x, x'; L)$, such that [14, 17]:

$$\left(-\frac{b}{4} (\partial_\alpha - iA_\alpha)^2 + \mu + i\phi \right) \tilde{G}(x, x'; \mu) = \delta^2(x - x') \quad (11)$$

The Laplace transformed Green function, $\tilde{G}(x, x'; \mu)$, can additionally be written as the two-point function of a complex field theory, derived from the following replicated Hamiltonian (see e.g. [11, 12, 14, 17] for related work):

$$H_n = \int d^2x \left[\frac{b}{4} \sum_{a=1}^n (D_\alpha \psi_a)^* (D^\alpha \psi_a) + \mu \sum_{a=1}^n \psi_a^* \psi_a + i\phi \sum_{a=1}^n \psi_a^* \psi_a \right] \quad (12)$$

with replica index a , and covariant derivative: $D_\alpha = \partial_\alpha - iA_\alpha$.

Following in the footsteps of [19], we now write for our replicated complex field variable, ψ_a : $\psi_a = \chi_a$, where χ_a is restricted to be a purely real field. In this way the ‘current’ term, $J_a^\alpha = i(\psi_a^* \partial^\alpha \psi_a - \psi_a \partial^\alpha \psi_a^*)$, appearing in Eq.(12) vanishes identically. Polymer states corresponding to nonzero values of such a current are thus disfavoured, due to a correspondingly higher energy cost (as analogously demonstrated in [19]).

Integrating out the auxiliary field ϕ , and the vector field A_α (utilising a large momentum cut-off π/a), we arrive in the appropriate, high density, limit of $\frac{b}{6} \sum_{a=1}^n \chi_a^2 \gg 1$ at (for related work see [11, 12, 19], for example):

$$H'_n = \int d^2x \left[\frac{b}{4} \sum_{a=1}^n (\partial_\alpha \chi_a)^2 + \mu \sum_{a=1}^n \chi_a^2 + \frac{\nu}{2} \left(\sum_{a=1}^n \chi_a^2 \right)^2 + \frac{\pi}{8a^2} \ln \left(\frac{eb}{6} \sum_{a=1}^n \chi_a^2 \right) \right] \quad (13)$$

RESULTS AND DISCUSSION

We now wish to calculate the ground-state energy of H'_n , corresponding to a condensed, confined, localised, and compact polymer configuration. Such high density polymer states are typically relatively weakly fluctuating, and in the thermodynamic (or large N) limit, tend to be well described by the principle of ground-state dominance [18, 20].

Following closely the work of [19], and invoking a type of ‘Lifshitz argument’ [20], we consider a slowly varying, radially symmetric, ground state given by the ansatz [19, 20]: $\chi_a = w_a J_0(j_{0,1} r/R)$. In this expression for χ_a , J_0 is the zeroth order Bessel function, and $j_{0,1}$ gives the location of its first zero ($j_{0,1} \approx 2.4$). Our variational ansatz is therefore simply described by just two parameters, namely its radius (or optimal size) R , and its amplitude w_a . Non-radially symmetric field configurations are more energetically costly, and hence can be safely ignored for the ground state [19].

It has been found that the appropriateness of such a ground-state trial-function for describing confined, or localised, states can be verified using the approach of [19]. The dominant contribution to the low-energy region, or lowest energy configuration, being well described by the large scale physics, or long wavelength mode, characterised by the overall size parameter R [19].

Note that, unlike the work of e.g. [11], we are not probing the (topologically induced) polymer collapse transition point in this work. Rather, we are investigating deep in the condensed polymer phase. Indeed, our approach has in some ways many similarities to that of placing a polymer chain in a rather deep, radially symmetric, potential well [18, 20]. There it is found again, using the well-known ‘Lifshitz argument’ [20], that the principle of ground-state

dominance typically leads to confinement, or localisation, of polymer phase behaviour corresponding to the lowest energy eigenvalue of an appropriately chosen eigenfunction [20] (satisfying certain boundary conditions [18]).

Furthermore, the Laplace variable μ , in a saddle point approximation, can be seen to merely serve to enforce the constraint: $\int d^2x \sum_{a=1}^n \chi_a^2 = L$. This constraint will ultimately allow us to normalise the amplitude of our ansatz, thus fixing the value of $\sum_{a=1}^n w_a^2$. Such a constraint is also broadly consistent with the closely related formal result (see e.g. [12]) for the polymer density in terms of replicated field variables: $\rho = \sum_{a=1}^n \chi_a^2$.

Inserting our variational ansatz into Eq.(13), we ultimately obtain:

$$H'_n = d_1 \frac{bL}{R^2} + d_2 \frac{\nu L^2}{R^2} + d_3 \frac{R^2}{a^2} \ln \left(d_4 \frac{bL}{R^2} \right) \quad (14)$$

where, for notational simplicity and convenience, we have introduced the following (purely numerical) constants, given by: $d_1 = \frac{c_2 j_{0,1}^2}{4c_1}$, $d_2 = \frac{c_3}{4\pi c_1^2}$, $d_3 = \frac{\pi^2}{8}$, $d_4 = \frac{e^{1+4c_4}}{12\pi c_1}$, and $c_1 = \int_0^1 u du J_0^2(j_{0,1} u)$, $c_2 = \int_0^1 u du J_1^2(j_{0,1} u)$, $c_3 = \int_0^1 u du J_0^4(j_{0,1} u)$, $c_4 = \int_0^1 u du \ln(J_0(j_{0,1} u))$.

The size dependence of our condensed polymer state can now be obtained by variationally minimising Eq.(14), w.r.t. the only remaining relevant collective coordinate R . In this way, we are estimating the ground-state energy as a function of the optimal size R . In the theory of disordered systems this type of approach has proven to be previously successful in estimating the size R of localised states, where it corresponds to (or is typically referred to as) the optimal-fluctuation method [19, 21].

Such an approach (albeit approximate) is very useful, since it permits us to investigate the long-distance properties of our polymer system in the non-perturbative regime. Moreover, it is precisely this long-distance physics which ultimately determines the overall polymer size R , which is what we seek in this work.

Using $L = Nb$, and simply evaluating the numerical value of $d_1/d_2 \simeq 1.8$ from the values defined above (involving additional, purely numerical, constants), we can distinguish two different regimes for our confined polymer, dependent on the strength of the excluded volume interaction ν . We find that:

Regime (i): For $\nu N \lesssim 1.8$, the first term on the l.h.s. of Eq.(14) dominates over the second term, and hence via balancing with the third term we obtain $R \sim N^{\frac{1}{4}} \ln^{-\frac{1}{4}} N$. This corresponds closely to the randomly branched polymer result ($R \sim N^{\frac{1}{4}}$), albeit with the additional presence of a weakly N dependent logarithmic term ($\ln^{-\frac{1}{4}} N$).

Regime (ii): For $\nu N \gtrsim 1.8$, the second term on the l.h.s. of Eq.(14) dominates over the first term, and hence via balancing with the third term we get $R \sim N^{\frac{1}{2}}$, which corresponds to canonical random walk statistics.

Reassuringly, we can check *a posteriori*, and for the sake of self-consistency, that the (large density) condition $\frac{b}{6} \sum_{a=1}^n \chi_a^2 \gg 1$, is indeed satisfied for both regimes studied above.

In this work, valid in the condensed, compact, and high density polymer phase, it therefore seems that the effect of increasing the excluded volume interaction is to cause the polymer chain to swell from a branched-like state to a random walk type configuration. Then again, this more swollen state could just as well be thought of as corresponding to that of a polymer globule in two dimensions, given that in some sense a random walk in 2D is equivalent to a globular state, since they both represent space-filling curves, with a fractal dimension equal to the ambient space dimension.

Heuristically [11], and very crudely, we can think of the branched-like polymer result obtained in this work as follows. The energy of an ideal chain confined in a hole (or potential well) of size R is roughly $\sim bL/R^2$, while the energy of a chain unentangled with a lattice of obstacles goes like $\sim R^2/a^2$. Balancing these two terms, via a Flory type argument [22], leads to an approximate polymer size of $R \sim N^{1/4}$, characteristic of randomly branched chains. The appearance of the additional, weakly N dependent, logarithmic factor ($\ln^{-\frac{1}{4}} N$) in our derived expression for the polymer size R given above, arises naturally due to the topological constraint, mediated by the gauge field A_α , as follows. Taking into proper account the topological interaction, via functionally integrating out the gauge field A_α in Eq.(12), introduces a concomitant, logarithmically dependent, contribution to the expression for the effective polymer Hamiltonian given by Eq.(13). Despite being presumably challenging numerically, it would be intriguing to see if the additional, weakly N dependent logarithmic correction factor, $\ln^{-\frac{1}{4}} N$, predicted in this work for the polymer size R could be potentially observed in any future simulation, or indeed experimental, work.

We should mention that, naturally, the results obtained in this work are necessarily specific to the 2D nature of the problem considered. It would be extremely interesting to try and extend our novel theoretical approach to the 3D case, including the use of the canonical Gauss linking number applied to a collection of mutually unentangled, self-avoiding, ring polymers in three dimensional space.

Finally, it is worth noting that the above arguments can be made rather more general [19], and that an analogous result could also be obtained by alternatively considering a random scalar field, coupled to the polymer in such a way as to give rise to a functional determinant, and hence an analogous $\sim R^2/a^2$ term in the free energy.

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FIGURES

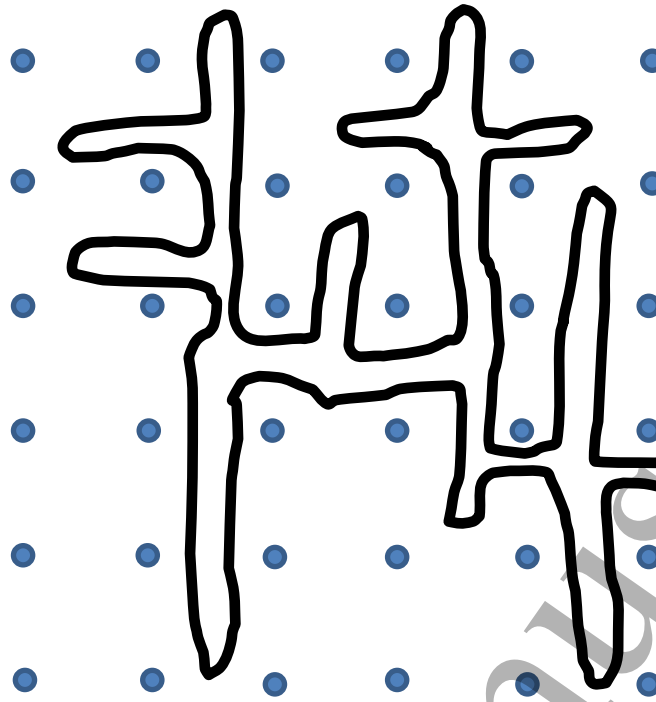


FIG. 1: Sketch of a 2D ring polymer, topologically unentangled with a planar lattice array of regularly spaced obstacles.