Ideal chains with fixed self-intersection rate

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We consider ideal chains in a hypercubic lattice $\mathbb{Z}^d$, $d \geqslant 3$, with a fixed ratio m of self-intersection per monomer. Despite the simplicity of the geometrical constraint, this model shows some interesting properties, such as a collapse transition for a critical value m_c. Numerical simulations show a self-avoiding-walk-like behavior for $m < m_c$, and a compact cluster configuration for $m > m_c$. The collapse seems to show the same characteristics as the canonical thermodynamical models for the coil-globule transition.

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I. INTRODUCTION

Polymer models have been the subject of extensive theoretical and numerical studies. Most of these models are based on ideal flexible chains, with the addition of various kinds of interactions between monomers, to include some nontrivial properties of real systems [1,2].

Very important is the *excluded volume* effect, which significantly modifies the fractal properties (the chains behave like self-avoiding walks, SAW); and the coil-globule (CG) transition, in which a flexible chain collapses from an extended coil to a liquid-like globule [2,3].

Most of the thermodynamic models that show the CG transition usually consider a competition between interactions of different geometrical nature. For instance, one can consider a lattice random chain with repulsive on-site interactions and attractive nearest-neighbor links. The transition arises from the competition of these interactions.

Let $\omega = \{S_0, S_1, \ldots, S_n\}$ be an n-step simple random walk (SRW) on the $\mathbb{Z}^d$ lattice (S_i are lattice vectors), and define the number of visits to each site x as $\psi_x = \sum_i \delta(x - S_i)$. A canonical model, incorporating excluded volume and a CG transition, is described by the Hamiltonian

$$\mathcal{H}[\psi] = \epsilon_0 \sum_x \psi_x^2 - \epsilon_1 \sum_{\langle x,y \rangle} \psi_x \psi_y, \tag{1}$$

where $\langle x,y \rangle$ indicates nearest neighbors (taking $\epsilon_1 = 0$ will lead to the Domb-Joyce model; see [4]).

A few years ago an interesting thermodynamic model that shows a transition with only on-site interactions was proposed [5]. However, a competition between two geometrically different constraints is still present since a self-avoidance is incorporated through restricting the maximal number of visits per site.

To our knowledge all the thermodynamic models proposed so far, having only one kind of short-range interaction, do not allow for the CG transition in the Boltzmann parameter.

In this work we will show how excluded volume effects, the CG transition, and liquid-like clusters can be obtained by imposing a single global geometric constraint. We consider ideal chains in a hypercubic lattice $\mathbb{Z}^d$, $d \geqslant 3$, with a fixed ratio m of self-intersections per monomer.

This model shows a CG transition for a critical ratio m_c: Numerical simulations (obtained by a standard implementation of the pruned-enriched Rosenbluth method, PERM, see [6–9]) indicate a SAW-like behavior for $m < m_c$ (for a review of SAW see [10–12]) and a cluster configuration for $m > m_c$. In addition the model is among the simplest with a crossover from SAW to cluster behavior. Our focus will be on $d \geqslant 3$ lattices because in these lattices, as we shall see, the transition occurs at nontrivial values of m_c.

II. MODEL DEFINITION

Consider an ideal chain ω of n steps on $\mathbb{Z}^d$ (without loss of generality we take $S_0 = 0$). We call $R[\omega]$ the range of ω (number of distinct lattice sites visited by the path), and $M[\omega] = (n+1) - R[\omega]$ the number of self-intersections.

Let $P_n(M)$ be the fraction of SRW, of length n, with exactly $M \in [0, n-1]$ self-intersections. We introduce mean value $\langle M_n \rangle$ and variance $\langle \Delta M_n^2 \rangle = \langle M_n^2 \rangle - \langle M_n \rangle^2$ of the distribution $P_n(M)$. It is well known (see [4]) that for SRW in $d \geqslant 3$, $d \neq 4$,

$$\langle M_n \rangle = C_d n - \Delta_d n^{2-d/2} + O(1), \tag{2}$$

where C_d is the probability that an infinite length walk contains its starting site at least twice (for numerical values of C_d, see [13]), and Δ_d is exactly known [for $d = 4$ the main fluctuation is actually $\Delta_4 \log(n)$] [4,13]. Concerning the variance, Jain and Pruitt have shown that $\langle \Delta M_n^2 \rangle \propto n \log(n)$ for $d = 3$, and $\langle \Delta M_n^2 \rangle \propto n$ for $d \geqslant 4$ [4,14]. They have also shown that for $d \geqslant 2$

$$\xi = \lim_{n \to \infty} (M - \langle M_n \rangle) / \left\langle \Delta M_n^2 \right\rangle \tag{3}$$

is normally distributed, from which it follows that $P_n(M)$ is peaked around its mean value for long walks.

We are interested in chains in which the rate of intersections per monomer is fixed at a certain value $m \in [0,1]$ in the thermodynamic limit. We define the ensemble Ω_m of n-step walks with exactly $M = \lfloor m(n-1) \rfloor$ intersections, and Ω, the ensemble of all n-step SRW; we call $\langle \cdot \rangle_m$ the average on Ω_m, and $\langle \cdot \rangle$ that of Ω.

For large n we can approximate $\lfloor m(n-1) \rfloor \simeq mn$. Henceforth we will work under this approximation. We introduce the fraction of m-intersection rate walks as $P_n(m)$. From the properties of $P_n(M)$ it follows that $P_n(m)$ is peaked around $\langle m \rangle = C_d$ when $n \to \infty$.

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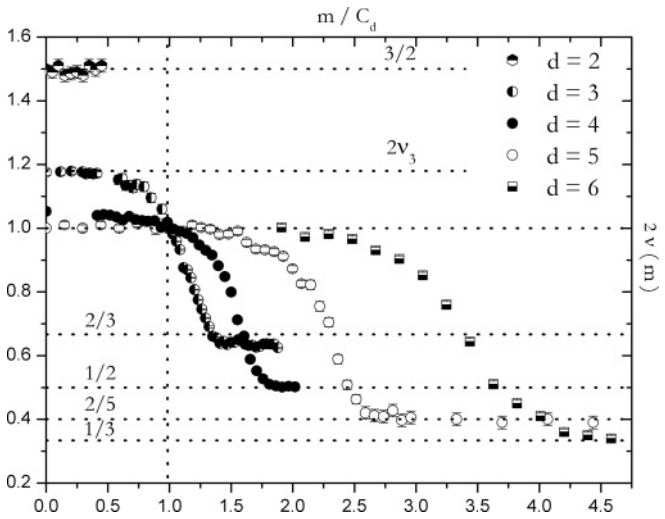

FIG. 1. Exponents $\nu_d(m)$ vs m/C_d from simulations at various lattice dimensions. The graph shows the behavior of $\nu_d(m)$, obtained from fits of $\log(\langle S_n^2\rangle_m)$ vs $\log(n)$. The range for all fits is $n \in [0.8n_M, n_M]$. Maximal length for simulated walks was $n_M = 2 \times 10^3$ for $d = 2, n_M = 10^3$ for $d = 3, n_M = 10^3$ for $d = 4, n_M = 0.5 \times 10^3$ for $d = 5$, and $n_M = 0.3 \times 10^3$ for $d = 6$. Moreover, for $d = 3$, the $\log(\langle S_n^2\rangle_m)$ has been achieved by using $m = C_3 M/\langle M_n\rangle$, with an n dependence, to take into account the finite size of the chains (see Fig. 2). For $d > 3$, the finite size effects are much weaker and this correction is not necessary. The picture for $d = 4$, $m < m_c$ shows an exponent slightly larger than 1 [a correction of order $O(10^{-2})$], consistent with the expected logarithmic correction (for chains of length 10^3 an exponent of $2\nu \simeq 1.070$ would be predicted). In the $n \to \infty$ limit, $\nu_d(m)$ is expected to be a step function (see Fig. 3). The present graph shows the step at m_c: For $d = 3{,}4$ we expect $m_c = C_d$; for $d \geqslant 5$ simulations suggest $m_c > C_d$.

Let $\langle S_n^2\rangle_m$ be the mean square end-to-end distance on Ω_m. From our simulations we find that the relation $\langle S_n^2\rangle_m \propto n^{2\nu_d(m)}$ holds for any dimension considered, with the exponent $\nu_d(m)$ dependent on m and d. Again, for $d \geqslant 3$, the simulations show the existence of a critical value $m_c \in (0, 1)$ beyond which the chains collapse into a compact liquid-like globule, with $\nu_d(m) = 1/d$. If instead $m < m_c$ we have the SAW-like behavior (Figs. 1 and 2; see also Fig. 3). As a preliminary observation we can state that, since for $m = 0$ we have the self-avoiding walk, then $\langle S_n^2\rangle_0 \simeq D_d n^{2\nu_d}$ (ν_d is the correlation length exponent for the SAW, see [10]). On the other hand, from Eq. (3) it follows that the relation

$$\int_{-b_n}^{b_n} d\epsilon \, \langle S_n^2\rangle_{C_d+\epsilon} P_n(C_d + \epsilon) \simeq n \tag{4}$$

should hold in the large n limit and $b_n = o(1)$. This is clearly confirmed by our simulations.

III. GENERAL RESULTS

We will briefly discuss the recurrent cases $d = 1{,}2$. For $d = 1$ we have a rather simple situation: In a linear lattice chain the end-to-end distance is proportional to the range, hence proportional to $n, \forall m < 1$. A unidimensional SRW is recurrent ($C_1 = 1$; see [4]); therefore no drop of $\nu_1(m)$ is expected for

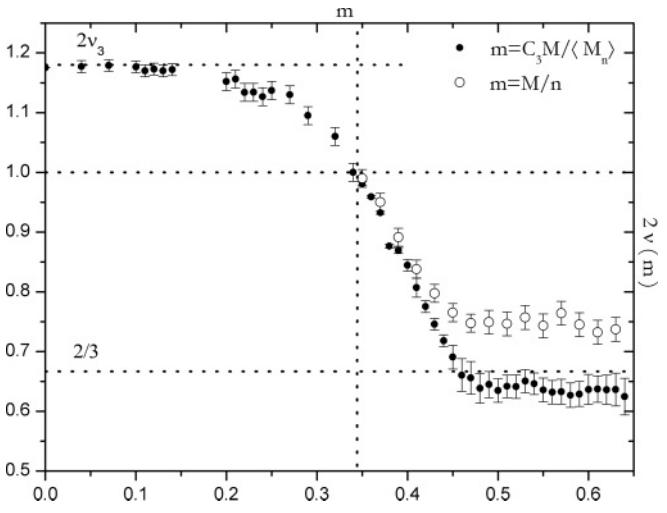

FIG. 2. Exponent $\nu_3(m)$ vs m, a comparison of simulations with and without finite-size corrections. The chart, from fits in the form $\log(\langle S_n^2\rangle_m)$ vs $\log(n)$, compares $m = M/n$ (empty circles) with $m = C_3 M/\langle M_n\rangle$ (full circles); $\langle M_n\rangle$ is obtained from Eq. (2). Essentially, M is rescaled with the average value of the support (range) $\langle R_n\rangle = n - \langle M_n\rangle$; this procedure leads to a significant improvement of the accuracy, at least for $m > C_3$ (vertical dotted line). Given that the two curves in the limit $n \to \infty$ must converge, it is clear that for $n_M = 10^3$ we are still far from the asymptotic regime. This fact is emphasized in Fig. 3 in which we can see how the drop band for $d = 3$ vanishes very slowly compared to $d > 3$.

$m < 1$. At $d = 2$ the picture is conceptually similar. We find $\nu_2(m) = \nu_2$ (data in Fig. 1), and since the square lattice SRW is still recurrent ($C_2 = 1$; from [4]) from Eq. (4) we expect that

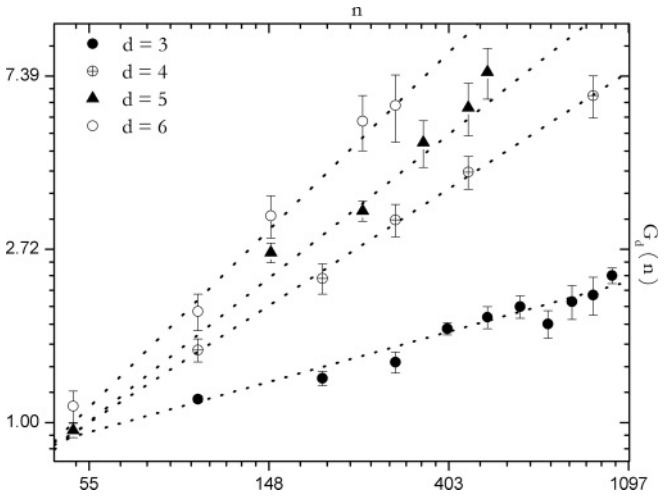

FIG. 3. Slope of $\nu_d(m)$ in the drop band. The graph shows (in log-log scale) the maximum slope $G_d(n) = -[\partial_m \nu_d(m)]_{\min}$ of $\nu_d(m)$ for different lengths n, which is an estimator of the *drop band* width [proportional to $G_d^{-1}(n)$] in which $\nu_d(m)$ falls from ν_d to $1/d$. $G_d(n)$ seems to increase with a power law, supporting the idea that $\nu_d(m)$ is a step function in the limit $n \to \infty$. Dotted lines are power law Bn^α fits to the data: $\alpha = 0.29 \pm 0.01$ for $d = 3$ (without the finite-size correction $C_3/\langle M_n\rangle$), $\alpha = 0.67 \pm 0.03$ for $d = 4$, $\alpha = 0.83 \pm 0.05$ for $d = 5$, and $\alpha = 1.02 \pm 0.07$ for $d = 6$. From this data we can notice that the width of the *drop band* decreases faster in n as lattice dimension d increases.

this relation holds for any $m < 1$ (see later discussion about the case $d = 3$).

The most important case is clearly $d = 3$: From simulations we find that $\nu_3(m < m_c) = \nu_3$, and $\nu_3(m > m_c) = 1/3$. The last statement needs some attention; we point out that $\nu_3(m > m_c) = 1/3$ is reached only after a long crossover. Figure 2 shows ν vs $n^{-1}M$ and $C_3 M/\langle M_n\rangle$. The former does not consider the finite size of the system, while the latter takes into account fluctuations of the walk mean support (range) $\langle R_n\rangle$ at finite n (see [15]). This enhancement allows a significant improvement of the accuracy for $d = 3$ [while is useless in higher dimensions, since, by Eq. (2), the range converges rapidly to its asymptotic behavior].

From Eq. (4) we can locate m_c for $d = 3$. If $\langle S_n^2\rangle_m$ has the SAW-like behavior (with $\nu_3 > 1/2$; see [11]) for $m < m_c$, and cluster-like for $m > m_c$, then the drop of $\nu_3(m)$ must lie within the range of integration of Eq. (4). It follows that $m_c = C_3$ for $n \to \infty$. As for the exponent at the critical point, Eq. (4) tells us that $\nu_d(m \to C_d) = 1/2$, from which it follows that $\nu_3(m_c) = 1/2$. This observation is supported by Figs. 1 and 2, where we see that $2\nu_3(m)$ passes through 1 at the expected critical point $m_c = C_3$.

This latter fact is of some importance, since the transition for $d = 3$ would show the same behavior as that described by the Hamiltonian in Eq. (1). If the two models belong to the same universality class this would be very interesting, since in our model the transition arises from the necessity of maximizing the configurational entropy, without the need for any further interactions. Indeed, at the critical point m_c there is a radical change in the optimal strategy to achieve the global constraint m (which is actually a long-range correlation). For $m < m_c$, the best way to change the ratio m is to compress (or expand) the chain locally, keeping the SAW-like fractal structure. In this situation the monomers intersect (on average) only within a certain distance along the chain. For $m > m_c$ instead, it becomes entropically convenient to assume a compact configuration, also allowing intersections between monomers very far apart in terms of position along the chain.

We also studied higher dimensions; the results are consistent with a CG transition of the same kind. For $d = 4$ we have $\nu_4(m > m_c) = 1/4$ and $\nu_4(m < m_c) = 1/2$, but the random-coil behavior shows logarithmic corrections. Simulations fit the conjecture that $\langle S_n^2\rangle_m \propto n \log(n)^{1/4}$, as in the SAW case [10]. If this is true, from Eq. (4), we should again find $m_c = C_4$.

As expected, for $d \geqslant 5$, $m < m_c$, we find the mean field behavior $\nu_d(m) = 1/2$ (as for the SRW), and $\nu_d(m) = 1/d$ for $m > m_c$. Since for $d \geqslant 5$ $\nu_d(m < m_c)$ is the same as for the SRW, we neither use Eq. (4) to locate m_c nor find $\nu_d(m_c)$, but our simulations [extremes of the drop zone of $\nu_d(m)$, $d = 5,6$ in Fig. 1] strongly suggest that $m_c > C_d$. All results and conjectures about the behavior of $\langle S_n^2\rangle_m$ have been summarized in Table I.

We would like to point out that all results for $d \geqslant 3$, $m \geqslant m_c$, are consistent with a remarkable exact work by van den Berg, Bolthausen, and den Hollander [16] on the moderate deviations for the volume of a Wiener sausage (WS), which is a neighborhood of the trace of a standard Brownian motion up to a time t, given by taking all points within a fixed distance a of Brownian motion (essentially a continuous version of our model; see [17]). Let $\eta(t)$, $t \geqslant 0$ be the standard Brownian motion in $\mathbb{R}^d$ starting at the origin. The WS $W^a(t)$ with radius a is the process defined by

$$W^a(t) = \bigcup_{0 \leqslant s \leqslant t} B_a(\eta(s)), \tag{5}$$

where $B_a(x)$ is the open ball with radius a around $x \in \mathbb{R}^d$. This paper considers the probability of having a Wiener sausage $W^a(t)$ of volume $|W^a(t)| \leqslant bt, b \in [0,\kappa_a]$ [$\kappa_a t$ is the long-time behavior of $\langle |W^a(t)|\rangle$], showing that there exists a critical value b_c below which the sausage is supposed to collapse in a *swiss-cheese* like compact configuration [a nonpercolating cluster with random holes of size $O(1)$]. They rigorously showed that $b_c \in (0,\kappa_d)$ for $d \geqslant 5$ only, while for $d = 3,4$ the transition is at $b_c = \kappa_a$ exactly.

TABLE I. Summary of predictions for $\nu_d(m)$. For $d = 5,6$ we were unable to look at the critical behavior, while the critical point m_c is evaluated by the drop zone of $\nu_d(m)$. The *l.c.* indicates SAW logarithmic correction in $d = 4$.

d	$\nu(m < m_c)$	$\nu(m = m_c)$	$\nu(m > m_c)$	m_c/C_d
3	ν_3	1/2	1/3	1
4	$1/2_{l.c.}$	1/2	1/4	1
5	1/2		1/5	$1.5 \sim 2.7$
6	1/2		1/6	$2.1 \sim 4.4$

IV. ORDER PARAMETER

Typically the order parameter considered for a CG transition is $n^{-1}\langle S_n^2\rangle_\beta$ (with $\langle\cdot\rangle_\beta$ being the thermodynamic average); we propose here a slightly different parameter:

$$\varrho_d(m) = \lim_{n\to\infty} \sqrt{\langle S_n^2\rangle_m / \langle S_n^2\rangle_0}. \tag{6}$$

This function vanishes beyond the critical point, similarly to the magnetization for spin systems. If the analogy with the CG transition from Eq. (1) holds, we should expect a second-order transition, at least for $d = 3$.

Our simulations (Fig. 4) do not allow us to clarify whether the transition is continuous at m_c. However, for $d = 3$ we can get some insights by a mean-field analysis of the Stanley model (SM; see [18]), a model of correlated random walks defined by the following partition function:

$$\mathcal{Z}_n(\beta) \propto \sum_{M=0}^{n} P_n(M) e^{-\beta M}. \tag{7}$$

Given that variable ξ in Eq. (3) is normally distributed, it is easy to show that for small β

$$\lim_{n\to\infty} n^{-1}\langle M_n\rangle_\beta \simeq C_3 - \delta m(\beta), \tag{8}$$

where $\langle M_n\rangle_\beta$ is the mean number of intersections in the SM at temperature β, and $\delta m(\beta)$ vanishes for $\beta \to 0$. From Eq. (3) we have a Gaussian shape $P_n(M)$ near $M = C_3 n$, with $\langle \Delta M_n^2\rangle \propto n \log(n)$. By replacing $M \to mn$ in Eq. (7) we find

$$\mathcal{Z}_n(\beta) \propto \int_0^1 dm\, \exp\left[-\frac{n}{2}\left(2\beta m + n\langle \Delta M_n^2\rangle^{-1} \delta m^2\right)\right] \tag{9}$$

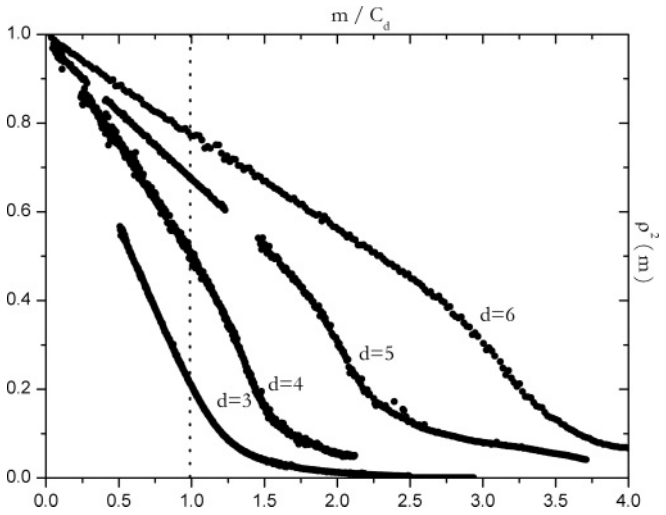

FIG. 4. Order parameter $\varrho_d(m)$ vs m/C_d from simulations at various lattice dimensions. The graph shows $\langle S_n^2\rangle_m/\langle S_n^2\rangle_0$: $n = 10^3$ for $d = 3, n = 2 \times 10^3$ for $d = 4, n = 10^3$ for $d = 5$, and $n = 0.5 \times 10^3$ for $d = 6$. The vertical dotted line is the expected transition point for $d = 3{,}4$ as $n \to \infty$. The asymptotic behavior near m_c is only hinted, since our PERM implementation, which requires us to store the microcanonical density of states at many n, has not allowed us to simulate longer chains. Better pictures could surely be achieved, by more focused techniques, for every dimensions except $d = 4$. For $d = 4$ and $m < C_4$ we expect $n^{-1}\langle S_n^2\rangle_m$ to be only $O(\log(n)^{1/4})$, implying the asymptotic behavior would be difficult to demonstrate clearly even by simulation for very large chains.

for small β, with $\delta m = (C_3 - m) \simeq 0$. Whereas there is a positive constant a_0 such that $\langle \Delta M_n^2\rangle > a_0 n$ for large n, performing a saddle point integration on Eq. (9) with $n \to \infty$ we obtain $\delta m(\beta) > c_0\beta$ (for small β and $c_0 > 0$).

From simple mean-field arguments (see [21]) it is reasonable to assume that the SM can be approximately described by the Flory theory. Consider the following Flory energy:

$$\mathcal{F}_\beta(r) \sim \beta n^2 r^{-d} + \frac{r^2}{2n} - (d-1)\log(r) + \Lambda(n), \quad (10)$$

where $r = |S_n|$ is the end-to-end distance, and $\Lambda(n)$ is a function independent of r [10]. We minimize the functional under the assumption $r \sim \beta^\theta n^\nu$; thus

$$d\beta^{1-\theta(1+d)} n^{2-\nu(1+d)} + (d-1)\beta^{-\theta} n^{-\nu} = \beta^\theta n^{\nu-1}. \quad (11)$$

When $\nu d < 2$ we find $\nu = 3/(d+2)$ and $\theta = 1/(d+2)$, otherwise $\nu = 1/2$ and $\theta = 0$. When $d = 3$ this theory predicts $\theta = 1/5$. A different estimation method uses path integrals; results are in agreement [22].

Flory theory for SM where $d = 3$ predicts $\langle S_n^2\rangle_\beta \propto \beta^{2/5} n^{6/5}$ for small β; inverting $\delta m(\beta)$, and substituting in the expression for $\langle S_n^2\rangle_\beta$, we find $\varrho_3(m) < b_0 \delta m^{1/5}$, with $\delta m = (C_3 - m) \simeq 0$, $b_0 > 0$.

In the case $d = 4$ Flory theory cannot be used, since the logarithmic correction to the mean-field behavior is not captured. Note that the $d \geqslant 5$ theory predicts $\theta = 0$, in agreement with the prediction $m_c > C_d$ for $d \geqslant 5$.

The exponents found are certainly incorrect, since $\nu_3 \neq 3/5$, but a power-law behavior $\varrho_3(m) \propto \delta m^\theta$ (with a likely logarithmic correction) is reasonable, suggesting that in $d = 3$ the transition is of the second order, with $m_c = C_3$.

V. CONCLUSION AND OUTLOOK

In this paper we introduced a new athermal model of interacting random walks, which shows a CG transition for a critical ratio between the range and the number of monomers. The transition seems to show the same characteristics as that seen in canonical models. However, the relationship between our microcanonical model and those canonical for the CG transition is nontrivial (for a general discussion on the differences between microcanonical and canonical ensembles see [23]).

The Hamiltonian in Eq. (1), as well as the interaction described in Ref. [5], operates a selection on the ensemble Ω whose mechanisms are not easily connected to the ensemble Ω_m. Although this is a key issue, we do not discuss it here; a work dedicated to this subject is currently under preparation, where we will present a detailed study of the $P_n(m)$ distribution and its relation to some thermodynamic models.

Apart from more focused implementation of the simulations here presented, other issues of interest could affect the connectivity properties of collapsed clusters (an example is found in [16]; collapsed WS clusters should be nonpercolating, with $O(n)$ holes sized $O(1)$ distributed inside the range; is reasonable to expect the same behavior for our model when $m > m_c$).

In general, this model certainly deserves attention since, in our opinion, it could lead to substantial improvements in understanding the geometry of CG transitions, as well as the crossover between the SAW and the SRW.

ACKNOWLEDGMENTS

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algorithm that combines the Rosenbluth-Rosenbluth method (which simulates a biased sample and corrects the bias by means of a weight associated with each configuration) with recursive enrichment, starting off building instances according to a biased distribution, but correcting for this by cloning desired (*enriching*) and killing undesired configurations (*pruning*) in order to contain the weights fluctuations of the simulated samples; see [7–9] for reviews and [7] for a pseudocode.

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[18] The Stanley model (SM, see [19] for the original formulation, or [4] for a review) is a model of correlated random walks which describes polymer chains with either repulsion or attractions. The attractive case is mathematically equivalent to the Rosenstock trapping model (random walk on lattice with randomly distributed irreversible traps: a walker encountering a trap is killed there; see [4]) and has been exactly solved by M. D. Donsker and S. R. S. Varadhan [20].

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[21] We show that for small β the SM is described by the same mean field theory of the Domb-Joyce model (see [4] for a review of this important model), therefore by the Flory theory. We start from the Domb-Joyce Hamiltonian $\mathcal{H}_{DJ} = \epsilon \sum_x \psi_x^2 = \epsilon \sum_x R_x \psi_x^2$, with $R_x = \lim_{\gamma\to 0} \psi_x^\gamma$ and ψ_x the same as Eq. (1): Approximating $\psi_x \to \bar{\psi} = nR^{-1}$ we have $\mathcal{H}_{DJ} \sim \epsilon R \bar{\psi}^2$, where $\bar{\psi}$ is the average occupation of a visited site, and $R = \sum_x R_x$ is the range. It follows that $\mathcal{H}_{DJ} \sim \epsilon n(1-m)^{-1}$, while the Hamiltonian of the SM is $\mathcal{H}_{SM} = -\epsilon nm$. For small β we have to consider $m = C_d - \delta m$, and substituting this expression in $\mathcal{H}_{DJ}$, $\mathcal{H}_{SM}$ we find $\delta\mathcal{H}_{DJ} \propto \delta\mathcal{H}_{SM}$, from which it follows that the two models are equivalent in mean-field description, with different couplings ϵ.

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