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Layering in crumpled sheets

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Abstract – We introduce a toy model of crumpled sheets, and use Monte Carlo simulation to show there is a first-order phase transition in the model, from a disordered dilute phase to a mixture with a layered phase. We demonstrate the transition through two order parameters, *corr* and *lay*, the first of which measures orientational order while the second measures bulk layering. An important feature of the argument is the behavior of the system as its size is increased.

Introduction. – When a sheet of stiff paper is crumpled into a compact ball, creases and folds appear. This storage of energy, especially in the irreversibly distorted creases, has been widely studied, for instance in [1–4]. Our interest here is in geometric changes associated with the (reversible) folds, which are less well understood.

Consider the densest possible state of the material, in which the stiff sheet is carefully layered into a compact stack of parallel leaves. We will be concerned with the transition between states of varying density. Imagine the process of compacting the sheet within a contracting sphere, from a typical initial state of low volume fraction near 0 to a typical state of high volume fraction near 1. These two regimes are noted for instance in [3,4]. Our paper focuses on whether as compaction proceeds, the connection between these extreme regimes is smooth. We in fact suggest that the connection is not smooth, but instead singular in a manner which is commonly called a *phase transition*, that is, a change in behavior at a precise degree of compaction in the infinite volume limit of the system.

Justification of this would need two components, theoretical and experimental. We discuss here mainly the theoretical aspect, through a model, but note some experimental issues in the last section.

We are interested in matter progressively confined as when a sheet is crumpled in one's hands. A sheet is thin in one of its dimensions. There is a natural aspect of this subject in which the material is thin in two of its dimensions, for instance compacted wires or linear polymers. In one sense these materials are simpler; sheets of paper cannot easily deform into a spherical cap, but instead form irreversible creases, while wires are not subject to this complication. For wires however there is a question of the manner of confinement; without any special constraint a wire could for instance produce a dense phase by coiling like a spring [5], which is irrelevant for sheets. To restrict to the essentials of both types of material we will discuss a model of a wire which is confined in a thin box as it is compacted, as has been done for instance in [6] where confining plates eliminate the possibility of coiling.

Crumpled materials are a form of soft matter, in the sense that they can be macroscopically deformed with much less energy than required to similarly deform a typical equilibrium solid. De Gennes [7], Flory [8], Edwards [9], and others have long championed the use of statistical mechanics methods to model various forms of soft matter, especially polymers, colloids and granular media. Recently, this approach has also been used to model sheets (see [4]), wires (see [10]) and polymers (see [5,11]) under variable confinement, even to model a possible phase transition, in the strict sense we are using, between the high- and low-density regimes. We note in particular two different types of modelling, a mean-field approach by Boué and Katzav [10] for a model of wires, and one using self-avoiding walks on a lattice by Jacobsen and Kondev [11] to model folded polymers. Both predict a second-order phase transition. We use a closely related approach but find a first-order phase transition, in contrast with the results of those papers. The difference is significant. Our results are evidence within this form of soft matter of an order/disorder transition with coexisting phases, known already in both the fluid/solid and liquid crystal transitions of equilibrium fluids, as well as in

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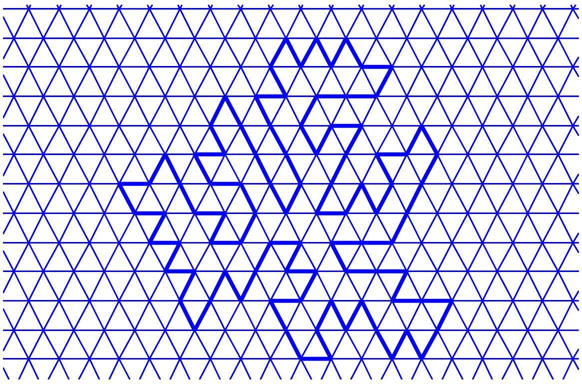

Fig. 1: (Color online) A self-avoiding polygon on L; orientation not indicated.

granular matter [12,13]. This suggests a basic, unified treatment of the phenomenon in a variety of materials both in and out of equilibrium.

In the last section we compare our results with previous work in [4,10,11] and discuss experimental verification.

The model and results. – For a fixed integer n consider the triangular lattice $L = \{(a+b/2, b\sqrt{3}/2)\colon (a,b) \in (\mathbb{Z}/n\mathbb{Z})^2\}$ with periodic boundary conditions. Note that this space is homogeneous and isotropic.

Let $\mathcal{A}$ be the set of oriented self-avoiding closed walks (polygons) on L, its elements being called walks or *configurations*; see fig. 1. If a walk $C \in \mathcal{A}$ changes direction at a vertex, we say there is a bend there. If it changes by $\pm 2\pi/3$, we call the bend large; if it changes by $\pm 2\pi/6$ we call it small. We define $B_s(C)$ as the number of small bends in C and $B_\ell(C)$ as the number of large bends in C. Assigning the energy e_s to each small bend and e_ℓ to each large bend, we define the total energy of configuration C by

$$\mathcal{E}(C) = B_s(C)e_s + B_\ell(C)e_\ell, \qquad (1)$$

and denote the model with energy parameters e_s and e_ℓ by $e_s : e_\ell$.

Because it is harder to simulate our model at fixed density and/or fixed energy, we use variables β to fix the average of energy $\mathcal{E}$, and μ to fix the average of particle (edge) number N, assigning the probability $m_\mu(C)$ for any $C \in \mathcal{A}$:

$$m_\mu(C) = \frac{e^{-\beta[\mathcal{E}(C)-\mu N(C)]}}{Z}, \qquad (2)$$

where Z is the normalization. (We suppress dependence on the system size.) Since we will only study isotherms and can choose the energies e_ℓ and e_s in the model, we fix $\beta = 1$ without loss of generality. We have simulated $1:2$ and $1:\infty$ with values of μ varied to examine a range of densities, and found the same qualitative results, which we present below.

To simulate the model at μ-values $\mu_0 < \mu_1 < \ldots < \mu_{l-1}$, we start with $\mu = \mu_0$ and a configuration which is a cycle with 6 edges. The end configuration in the simulation of μ_i is taken as the starting configuration in the simulation of μ_{i+1}. The basic Monte Carlo step is the following. From a given configuration $C \in \mathcal{A}$, we choose a random subpath σ_d of length d. We then consider all configurations $C' \in \mathcal{A}$ obtainable from C by replacing σ_d with a path $\sigma_{d'}$ while leaving the rest of C unchanged. Here σ_d and $\sigma_{d'}$ have the same starting points and ending points, and the length d' of $\sigma_{d'}$ is bounded, $d_1 \leqslant d' \leqslant d_2$. We then choose $C' \in \mathcal{A}$ with relative acceptance probabilities determined by m_{μ_i}. For the $1:2$ model we choose, with probability 0.5, either $d = 1$ and $d_1 = d_2 = 2$, or $d = 2$ and $d_1 = d_2 = 1$. For the $1:\infty$ model we take $d = 5$, $d_1 = 1$ and $d_2 = 9$.

For each measurement *meas* we use a standard autocorrelation function to find a "mixing time" at each μ_i, which we define as the smallest value of t such that the autocorrelation

$$\frac{1}{(m-t)\sigma^2}\sum_{i=1}^{m-t}(meas(C_i)-\lambda)\cdot(meas(C_{i+t})-\lambda) \qquad (3)$$

falls below zero. Here $C_1, \ldots, C_m$ is the Monte Carlo chain of configurations corresponding to the simulation of μ_i, and λ and σ^2 are the (sample) average and variance of *meas* over that chain. We found that for each of our measurements *meas* (described below), our simulations of each μ_i were on average at least 20 times as long as the corresponding mixing times. We therefore believe our simulations sample the target distribution given by eq. (2) at each μ_i. We obtain error bars from the Student's t-distribution by running 200 independent copies of the simulation.

The infinite volume limit is important in our considerations, since there cannot be a transition in a model of finite size. In our simulations we approximate the infinite volume limit by measuring "bulk" effects, that is, features which scale with the size of the system. We introduce two such measurements, *lay* and *corr*, to detect the spontaneous symmetry breaking and layering which may occur at large μ. To detect "bulk" edge correlation, we define $corr(C)$ as follows: choose a random edge in C and define $corr(C)$ as the proportion of edges in C which are parallel to it. Since the model is isotropic we expect $corr(\mu)$ to be identically $1/3$ for small μ in the infinite volume limit.

To detect "bulk" layering we define $lay(C)$ as the size of the largest parallelogram centered at the origin in C which contains an 80% perfectly layered region; $lay(C)$ is then normalized by the system size. (A perfectly layered region consists of parallel line segments with no gaps or bends.) We expect that for small μ, *lay* is identically zero in the infinite volume limit. Note that the choice of 80% is rather arbitrary; any percentage significantly above 33% should detect bulk layers.

The data, from systems of volume $40^2 = 1600, 60^2 = 3600, 80^2 = 6400$ and $100^2 = 10000$, gives strong evidence that in the infinite volume limit $corr(\mu)$ is identically $1/3$ for $\mu < \mu^*$, and that $corr(\mu) > 1/3$ for $\mu > \mu^*$, where $\mu^* \approx 0.63$ for the $1:2$ model and $\mu^* \approx -0.2$ for the $1:\infty$ model. Furthermore the data strongly suggests that *lay* is

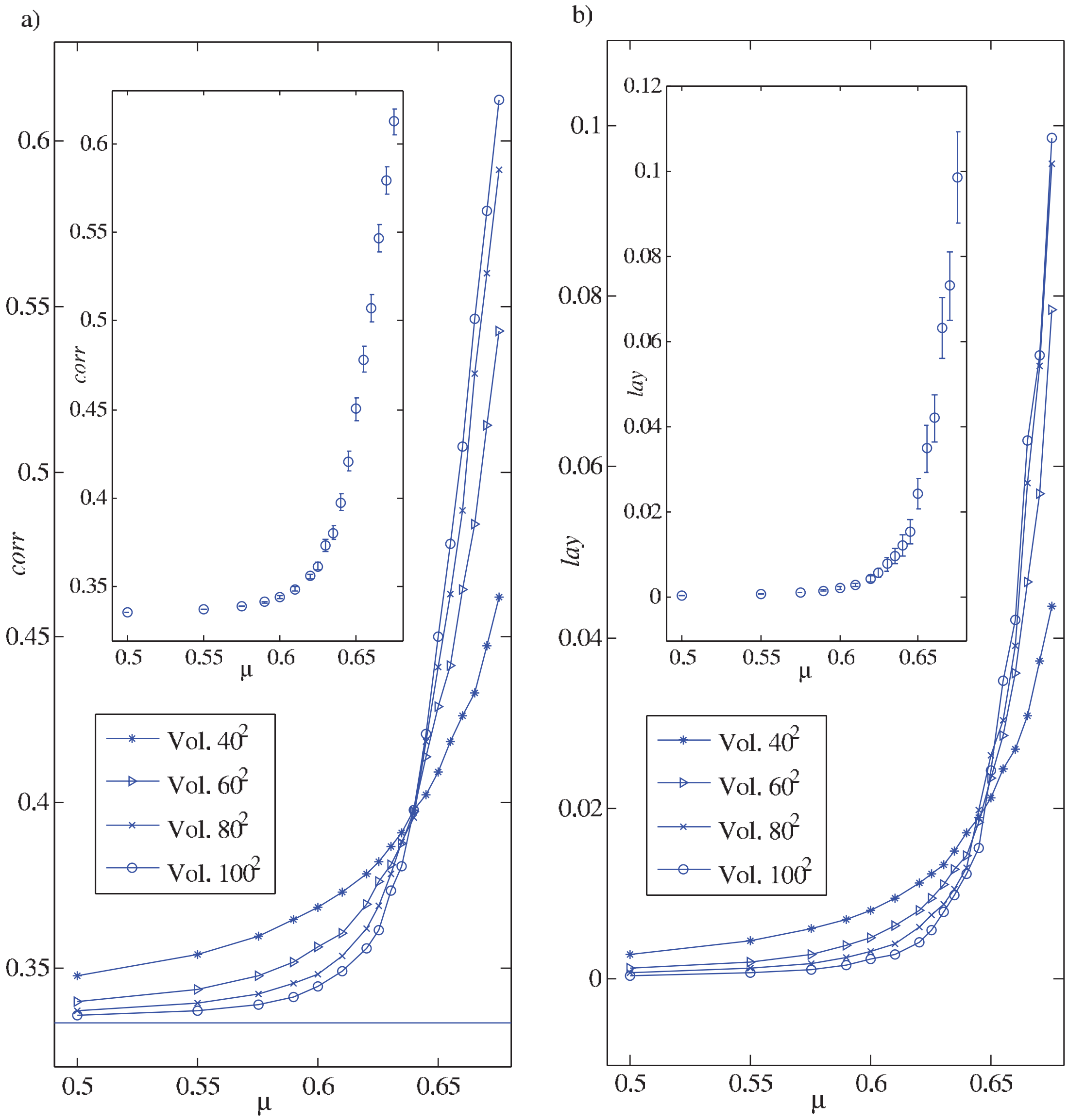

Fig. 2: (Color online) Data from the 1 : 2 model. a) Correlation *vs.* μ, for volumes 40^2 through 100^2, where the horizontal line represents $corr = 1/3$; b) layer size *vs.* μ, for volumes 40^2 through 100^2. Error bars in both insets represent 95% confidence intervals for volume 100^2, and for low μ are smaller than the data circles.

identically zero below μ^*, but positive above μ^*, showing the emergence of bulk layers above μ^*. See figs. 2, 3 and 4, 5, 6 with error bars in the insets.

Our argument for a phase transition is not based on any visibly developing kink in the graphs of the order parameters *corr* and *lay*. Instead, we argue that *in the infinite system* our order parameters, *lay* and *corr*, are identically 0 and 1/3, respectively, at low μ (indicating disorder), but rise above these values at high μ. We conclude that *lay* and *corr* are not analytic, since an analytic function which is constant on an interval is constant everywhere. Because *corr* and *lay* capture relevant physical information about the system we can reasonably call this a phase transition.

The rise of *lay* above zero indicates that there are coexisting layered and disordered phases; this is why we call the transition first order. Such a transition would imply a discontinuity in $\phi(\mu)$ (see fig. 7). We note that the graph of $\phi(\mu)$ does not clearly show a developing discontinuity; however, this is expected since the systems are finite and the plotted values of μ do not extend above the coexistence region. For this reason our argument for a first-order phase transition relies solely on the behavior of *corr* and *lay*.

Discussion of results. – We have introduced and simulated a toy model for the folding of a wire which is held in a thin box as it is progressively compacted. In the model, bulk folding begins to emerge at a sharp volume fraction as the material is compacted, just as, experimentally, bulk solid begins to emerge at a sharp volume fraction in the freezing transition of equilibrium

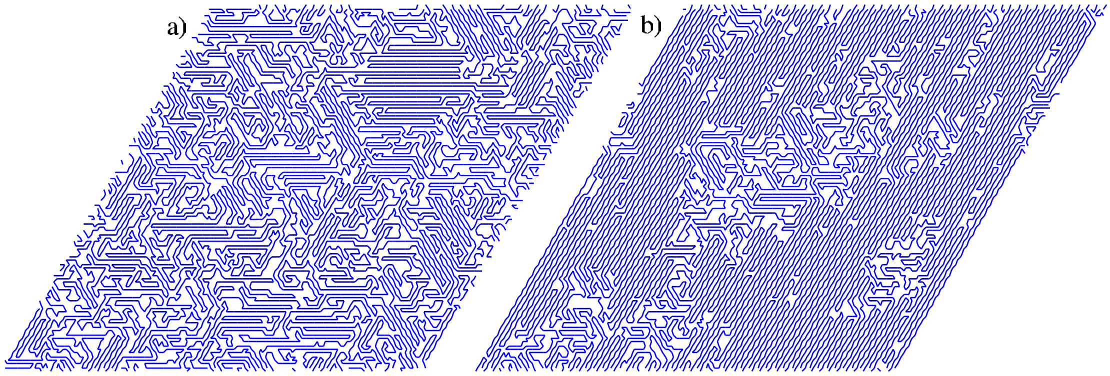

Fig. 3: (Color online) Data from the $1:2$ model. Snapshots of a loop in available volume 100^2 in equilibrium at $\mu = 0.6$ in a), and $\mu = 0.67$ in b).

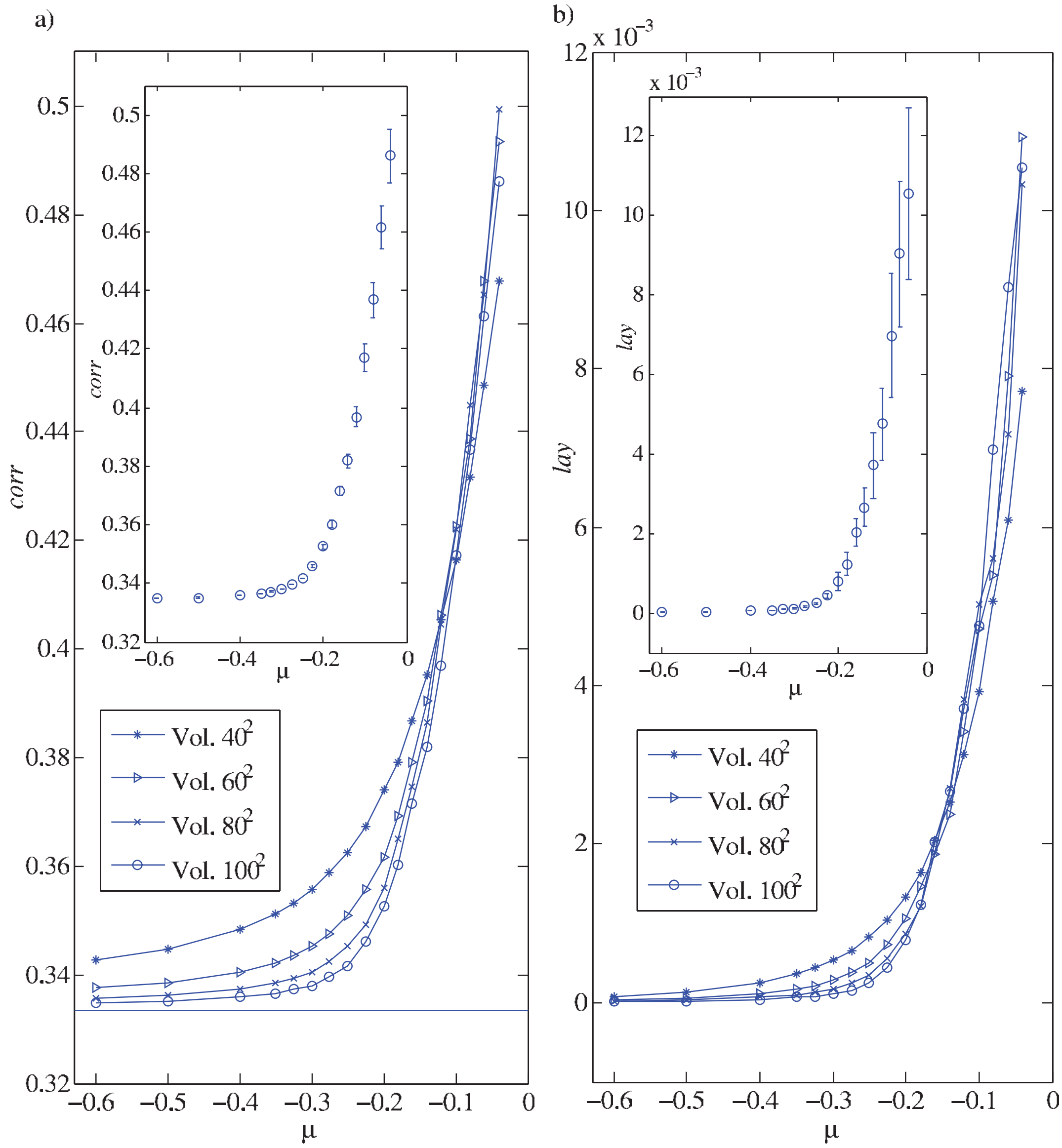

Fig. 4: (Color online) Data from the $1:\infty$ model. a) Correlation *vs.* μ, for volumes 40^2 through 100^2, where the horizontal line represents $corr = 1/3$; b) layer size *vs.* μ, for volumes 40^2 through 100^2. Error bars in both insets represent 95% confidence intervals for volume 100^2, and for low μ are smaller than the data circles.

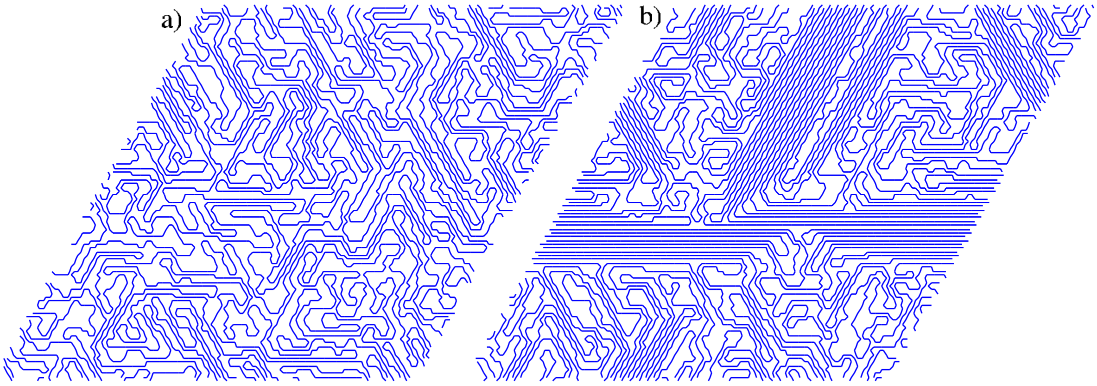

Fig. 5: (Color online) Data from the $1:\infty$ model. Snapshots of a loop in available volume 100^2 in equilibrium at $\mu=-0.3$ in a), and $\mu=-0.1$ in b).

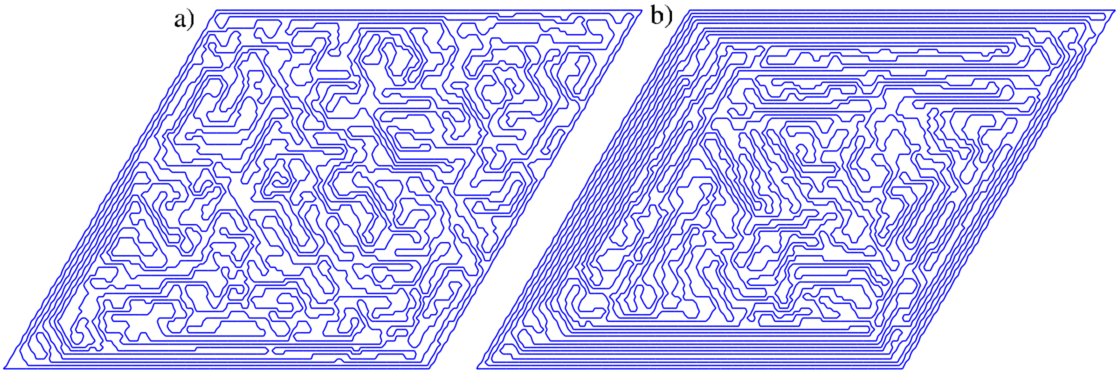

Fig. 6: (Color online) Data from the $1:\infty$ model with hard-wall boundary conditions. Snapshots of a loop in available volume 100^2 in equilibrium at $\mu=-0.3$ in a), and $\mu=-0.1$ in b).

fluids. This analogy has previously been used to model the behavior of other types of soft matter, in particular colloids [14] and the random close packing of granular matter [12,13].

As noted in the Introduction this paper is only one in a long history of modelling soft matter, not in thermal equilibrium, by the methods of equilibrium statistical mechanics. Such efforts are sometimes justified by assuming an external energy source, perhaps a vibration of the material, which might enable the microstate of the material to effectively optimize within the ensemble.

We next compare our model to three others. In [11] Jacobsen and Kondev analyze the Flory model [15] of strongly confined polymers using self avoiding walks on a 2-dimensional square lattice. Their model only considers walks which fill the confining region, that is, they are all of density one, but the model incorporates a parameter, which might be thought of as temperature, by which the ensemble average of the energy of the walks can be varied. They find a 2-dimensional "melting" phase transition by varying the temperature, or, equivalently, the strength of the interparticle interaction.

It becomes progressively more difficult in our model to simulate configurations of increasing density, so our results concern behavior as density increases starting from the low end. In order to produce bulk folding with increased density we needed to enforce low energy through the strength of the interaction. We do not vary this strength; in comparison with the Flory model, our sole variable is the density, or equivalently the chemical potential, and we move the system on a (low-temperature) isotherm. So we are in a sense exploring low-energy microstates of variable density but starting from the low-density end, and we therefore report our results as showing a "freezing" transition. It is possible that our first-order freezing transition is compatible with the second-order melting transition of [11]; see part C of sect. VII in [11].

We note that Boué and Katzav, in their model of confined wires [10], also obtain a second-order transition. They note the possible significance of their mean-field approximation and suggest Monte Carlo simulation as a way of eliminating need of the approximation. We feel the most likely source of our different conclusions is that their analysis leads them to the second-order transition at density 1 noted above, though we cannot rule out the effect of our use of a lattice.

In [4] Sultan and Boudaoud introduce a toy model for crumpled sheets, not wires, which however uses random

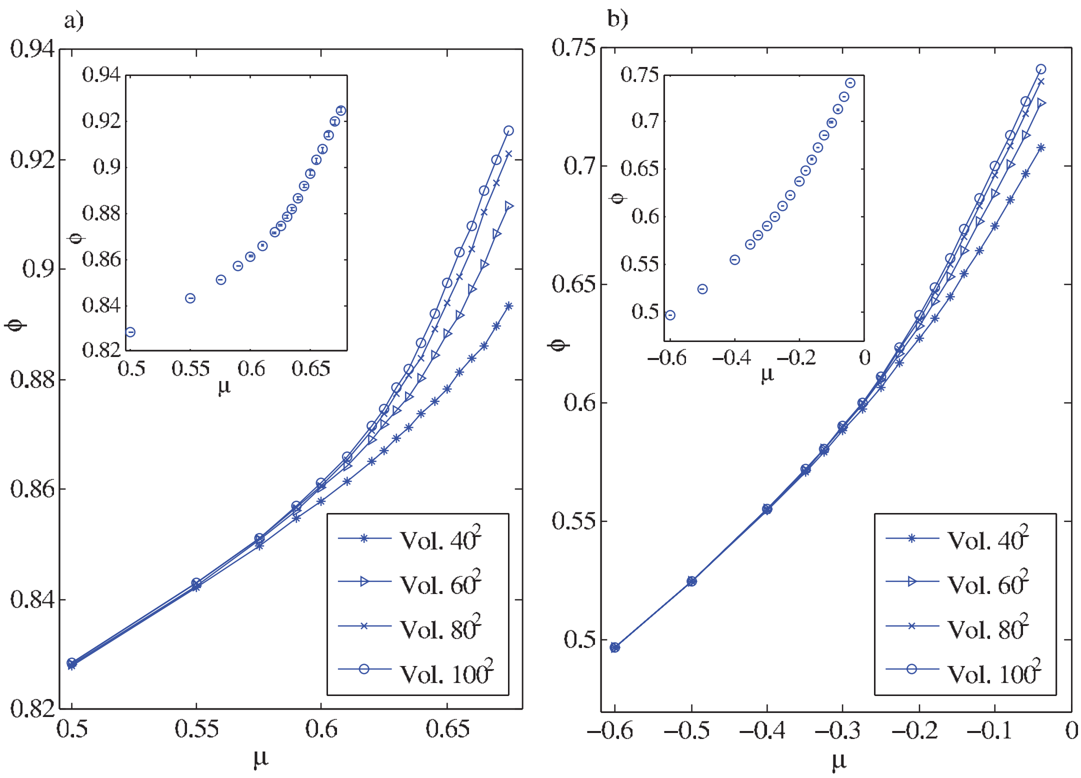

Fig. 7: (Color online) Volume fraction *vs.* μ, for volumes 40^2 through 100^2, for model $1:2$ in a) and model $1:\infty$ in b). Error bars in the inset figure represent 95% confidence intervals for volume 100^2, and are mostly smaller than the data circles.

walks in the plane in an interesting way. As in our model they attribute an energy to each walk and have only one parameter, controlling the compaction. In contrast their ensemble consists only of energy minima, and they investigate the "jamming" of the random walk under increasing compaction. They do find a qualitative difference between the states of low and high density, but there does not seem to be a claim of a sharp phase transition in our sense.

Our results explore a different regime (lower density) than the above analyses, with a somewhat different technique, and arrive at a significantly different conclusion. We feel it is particularly apt to explore all reasonable approaches in this subject since there is so much unknown at a basic level. Our results could be tested by confining a thin loop of wire held between parallel plates, and measuring the characteristics of the folding, in the two senses measured in our simulations, which emerge as the loop is progressively compacted. We are thinking of a physical setup as in [6]. Similar quantities could in principle be measured in confined sheets but it might be harder to analyze the folds in such an experiment.

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