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LETTER TO THE EDITOR

Folding of the triangular lattice in the face-centred cubic lattice with quenched random spontaneous curvature

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Received 29 August 1997

Abstract. We study the folding of the regular two-dimensional triangular lattice embedded in the regular three-dimensional face centered cubic lattice, in the presence of quenched random spontaneous curvature. We consider two types of quenched randomness: (1) a 'physical' randomness arising from a prior random folding of the lattice, creating a preferred spontaneous curvature on the bonds; (2) a simple randomness where the spontaneous curvature is chosen at random independently on each bond. We study the folding transitions of the two models within the hexagon approximation of the cluster variation method. Depending on the type of randomness, the system shows different behaviours. We finally discuss a Hopfield-like model as an extension of the physical randomness problem to account for the case where several different configurations are stored in the prior prefolding process.

The statistical properties of polymerized membranes have been extensively studied in the past few years [1, 2]. Some particular attention was paid to the role of quenched disorder in the elasticity of the membrane, with mainly two motivations. The first one is to understand the mechanism of the 'wrinkling' transition of partially polymerized lipid vesicles [3]. Such membranes undergo a reversible phase transition from a high-temperature soft phase with strong fluctuations to a low-temperature rigid and highly wrinkled phase. A second motivation is, at a macroscopic level, the study of the statistical properties of randomly crumpled paper or, more generally, randomly crumpled elastic sheets [4, 5]. In a random crumpling process, creases are created, which generate random spontaneous curvature. When iterated, the random crumpling processes can moreover cause frustration, and the crumpled paper may then have many equally probable configurations of minimal energy, a usual characteristics of random spin systems.

Here we study a simple system of a two-dimensional polymerized object with quenched disorder. As a toy model, we consider the problem of *folding* of the regular two-dimensional triangular lattice in the presence of random spontaneous curvature. Models of folding have been introduced in [6] and studied in [7, 8]. Originally, the study was restricted to *planar folding*, i.e folding in a two-dimensional embedding space. A more general discrete folding model with a three-dimensional embedding space was then introduced and studied in [9–11], describing the folding of the triangular lattice in the regular three-dimensional face-centred

0305-4470/97/240829+10\$19.50 © 1997 IOP Publishing Ltd

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cubic (FCC) lattice. The role of disorder in the *planar folding* problem was analyzed by the authors and Di Francesco in [12]. There, disorder was introduced in the form of a quenched random bending rigidity. Here we would like to complete our study by considering the folding in the three-dimensional FCC lattice in the presence of *quenched random spontaneous curvature*.

Before we proceed to our study, let us recall the results of [12] for the planar folding problem with random bending rigidity. There, we were interested in modelling the randomness arising from a prior irreversible 'crumpling' of the lattice. Such a crumpling results in a marking of the lattice bonds with quenched creases on which folds are favoured. The system can then be described by a Mattis-like model [13] with Hamiltonian

$$\mathcal{H}_{\text{Mattis}} = -K \sum_{\text{n.n.}(ij)} \tau_i \tau_j \sigma_i \sigma_j \tag{1}$$

where the variables $\sigma_i = \pm 1$ describe the (up or down) normal to the triangle *i* in the folded configuration and n.n.(*ij*) means summation over all nearest neighbour pairs. The disorder variables $\tau_i = \pm 1$, accounting for the prior irreversible marking, define a random bending rigidity $K_{ij} = K \tau_i \tau_j$, and are 'frozen' according to a specified probability distribution [12]. For the variables σ to represent actual folded configurations of the lattice, the six neighbouring spins on an elementary hexagon of the lattice, σ_i (i = 1, 2, ..., 6), must satisfy the 'physical' constraints [6–8]: $\sum_{i=1}^{6} \sigma_i = 0 \mod 3$. This condition, in particular, prevents one from absorbing the disorder in a simple change of σ_i into $\sigma_i \tau_i$. Similarly, if the corresponding disorder variables τ_i (i = 1, 2, ..., 6) arise from a prefolding process, they should also obey the physical rule $\sum_{i=1}^{6} \tau_i = 0 \mod 3$. This condition on the τ variables is essential for the system to develop a large K 'frozen phase' where the prior irreversible folded shape is recovered. If the τ variables are free ± 1 variables which do not satisfy the physical constraint above, the system becomes frustrated and the lattice remains in a disordered phase.

We now would like to extend these results to the case of the three-dimensional FCC folding problem. A folding of the triangular lattice in the FCC lattice is simply a mapping sending each vertex of the triangular lattice onto a vertex of the FCC lattice, with the requirement that neighbouring vertices on the triangular lattice remain nearest neighbours in the FCC lattice [9, 10], i.e. belong to the same triangular face. The FCC lattice is indeed made of octahedra and tetrahedra in contact by their triangular faces. Elementary triangles of the triangular lattice are thus sent onto elementary triangular faces of the FCC lattice. In the folded configuration, two adjacent triangles can form some relative angle θ , with one of the four following values:

(i) $\theta = 180^{\circ}$, no fold: the triangles are side by side;

(ii) $\theta = 0^{\circ}$, complete fold: the triangles are on top of each other;

(iii) $\theta = \arccos(1/3) \sim 71^\circ$, fold with acute angle: the two triangles lie on two adjacent faces of the same tetrahedron in the FCC lattice; and

(iv) $\theta = \arccos(-1/3) \sim 109^\circ$, fold with obtuse angle: the triangles lie on two adjacent faces of the same octahedron in the FCC lattice.

It was shown in [9] that these four types of folds can be understood as the superposition of the domain walls of two Z_2 variables $\sigma = \pm 1$ and $z = \pm 1$ living on the faces of the triangular lattice. The relative values $\Delta \sigma \equiv \sigma_2 \sigma_1$ and $\Delta z = z_1 z_2$ for two neighbouring triangles indicate which type of fold they form, with the correspondence displayed in table 1.

In order to describe an actual allowed folded state, the σ and z variables are subject to two basic folding rules involving the values σ_i and z_i (i = 1, ..., 6) on the six neighbouring triangles forming an elementary hexagon in the lattice. For each hexagon, the variables σ

Table 1. The relative folding state of two neighbouring triangles according to the relative values $\Delta \sigma$ and Δz .

$\Delta \sigma$	Δz	θ	Angle
1	1	180°	No fold
$^{-1}$	1	0°	Complete fold
1	$^{-1}$	71°	Acute field
-1	-1	109°	Obtuse fold

must satisfy the following first folding rule:

$$\sum_{i=1}^{6} \sigma_i = 0 \mod 3.$$
 (2)

This rule is identical to the rule of planar folding [7] although its interpretation here is slightly different [9, 10]. A second basic folding rule involves both the z and σ variables and reads

$$\prod_{i \in I(c)} z_i z_{i+1} = 1 \qquad \text{for } c = 0, 1, 2 \qquad I(c) = \left\{ i : \sum_{k=1}^l \sigma_k = c \mod 3 \right\}.$$
(3)

With the 'physical' constraints (2) and (3), one finds exactly 96 possible hexagonal configurations for the six triangles surrounding any of the vertices of the triangular lattice. Note that the planar folding problem can be recovered by freezing the z variable to $z_i = +1$ globally for all triangles. One is then left with exactly 11 possible hexagonal configurations.

In the absence of disorder, the folding energy is simply $E_{pure} = -K \cos(\theta)$ per lattice bond, with K the bending rigidity parameter. In terms of the variables σ_1, σ_2 and z_1, z_2 of the two triangles forming the fold, the folding energy simply reads

$$E_{\rm pure} = -\frac{K}{3}\sigma_1\sigma_2(1+2z_1z_2).$$
 (4)

The total folding energy is the sum of all elementary folding energies for all the bonds of the triangular lattice.

Disorder can be put in the model by introducing quenched disorder variables τ_i and w_i describing the prefolded state created by the irreversible crumpling process. The domain walls for the variables τ and w encode the four possible types of created creases with angle $\psi = 180^\circ$, 0° , 71° or 109° according to a table similar to table 1. Of course, in order to describe an actual prefolded state, the τ and w variables are subject to two local folding rules similar to (2) and (3). The presence of random creases directly leads to a random spontaneous curvature in the system, encoded in the angle ψ of the crease. Given this angle, the energy becomes minimum when the angle θ of the fold is such that $\theta = \psi$. We shall thus consider the following bending energy:

$$E = -K\cos(\theta - \psi) \tag{5}$$

where *K* measures the strength of this bending energy and where the quenched random variable ψ describes the quenched random spontaneous curvature in the system. As mentioned above, the variables θ and ψ take four values, leading to 16 possible fold/crease configurations. We would like to express the energy (5) in terms of the spin variables σ_i , z_i , τ_i and w_i (i = 1, 2) on the two neighbouring triangles on each side of the fold, as we did

in (4) for the pure case without disorder. The relative values $\Delta \sigma \equiv \sigma_1 \sigma_2$ and $\Delta z \equiv z_1 z_2$, on the one hand, and those of $\Delta \tau \equiv \tau_1 \tau_2$ and $\Delta w \equiv w_1 w_2$, on the other, fix the angles θ and ψ and thus the energy (5).

We can make use of the symmetry $\theta \leftrightarrow \psi$ in (5) (i.e. $(\Delta\sigma, \Delta z) \leftrightarrow (\Delta\tau, \Delta w)$) and of the symmetry $(\theta, \psi) \leftrightarrow (180^{\circ} - \theta, 180^{\circ} - \psi)$ (i.e. $(\Delta\sigma, \Delta\tau) \leftrightarrow -(\Delta\sigma, \Delta\tau)$) to ensure that the bending energy has the two *independent* symmetries $\Delta\tau \leftrightarrow \Delta\sigma$ and $\Delta w \leftrightarrow \Delta z$, and is even in $(\Delta\sigma + \Delta\tau)/2$, leading to the general form (for Z_2 variables)

$$\frac{E}{K} = e + a \frac{(\Delta z + \Delta w)}{2} + b_1(\Delta \sigma \Delta \tau) + b_2(\Delta z \Delta w) + c(\Delta \sigma \Delta \tau) \frac{(\Delta z + \Delta w)}{2} + d(\Delta \sigma \Delta \tau)(\Delta z \Delta w)$$
(6)

involving six constant coefficients e, a, b_1, b_2, c, d to be determined hereafter. These coefficients are simply obtained from the values of the six independent (i.e. not related by the above symmetries) pairs of folding and disorder configurations given in table 2.

Table 2. The folding and crease state of two neighbouring triangles and the corresponding bending energy/K for the six independent configurations.

Fold configuration	Disorder configuration	Bending energy/K
No fold	No crease	-1
No fold	Acute crease	1/3
No fold	Complete crease	1
No fold	Obtuse crease	-1/3
Acute fold	Acute crease	-1
Acute fold	Obtuse crease	-7/9

From this table, we determine the bending energy to be

$$E = \frac{K}{9} \left[-2 + 4 \frac{(\Delta z + \Delta w)}{2} - (\Delta \sigma \Delta \tau) - 2(\Delta z \Delta w) -4(\Delta \sigma \Delta \tau) \frac{(\Delta z + \Delta w)}{2} - 4(\Delta \sigma \Delta \tau)(\Delta z \Delta w) \right]$$
$$= -\frac{K}{9} [\Delta \sigma (1 + 2\Delta z) \Delta \tau (1 + 2\Delta w) + 2(1 - \Delta z)(1 - \Delta w)]. \tag{7}$$

The total bending energy is again the sum of all elementary folding energies for all links of the triangular lattice. It is interesting to check several limiting cases of the above formula. The pure folding problem, without disorder, can be recovered by constraining the disorder variables according to $\Delta \tau = \Delta w = 1$. The bending energy (7) reduces to $E = -K\Delta\sigma(1 + 2\Delta z)/3$, i.e to equation (4). The case of planar folding with disorder is obtained by setting $\Delta z = \Delta w = 1$ in (7) and the bending energy reduces then to $E = -K\Delta\tau\Delta\sigma$, as in (1).

In addition to the bending energy (7), we also introduce an external field H_r associated with the variable ($\sigma \tau + wz$), which is a rough measure of how close the folding configuration (σ , z) is from the disorder configuration (τ , w). This definition of the external field is not canonical and many other definitions are equally acceptable. The main motivation for introducing this external field is technical, i.e. the necessity to first prepare a solution with explicit broken symmetry to be able to eventually reach a solution with spontaneous symmetry breaking by tuning H_r to zero. Our total Hamiltonian is thus given by

$$\mathcal{H} = -\frac{K}{9} \sum_{\text{n.n.}(ij)} [\tau_i \tau_j (1 + 2w_i w_j) \sigma_i \sigma_j (1 + 2z_i z_j) + 2(1 - w_i w_j) (1 - z_i z_j)] -H_r \sum_i (\sigma_i \tau_i + z_i w_i).$$
(8)

To analyse the properties of the system, we will consider different order parameters. In view of what we know about the pure system without disorder, it is useful to divide the original triangular lattice into the two subsets A and B made of all triangles pointing up and all triangles pointing down, respectively, in the original flat triangular lattice. This, in particular, allows us to define 'staggered' average values. We will be interested primarily in the four following average values:

$$S_{\rm A} \equiv \overline{\langle \sigma \rangle_{\rm A}} \qquad Z_{\rm A} \equiv \overline{\langle z \rangle_{\rm A}} \qquad F_1 \equiv \overline{\langle \sigma \tau \rangle} \qquad F_2 \equiv \overline{\langle z w \rangle} \qquad (9)$$

where the brackets denote the average over the configurations at fixed disorder for an arbitrary given triangle (taken, moreover, in the subset A when we add the index A to the brackets) and the overline denotes the average over the quenched disorder. One has of course $-1 \leq S_A$, Z_A , F_1 , $F_2 \leq 1$. Non-zero values of the 'frozen' order parameters F_1 and F_2 indicate that the membrane is trapped in the configuration given by the disorder variables τ and w. The reason why we restrict ourselves to the subset A in S_A and Z_A is that, for the 'pure' system, that is the model without disorder, and at K = 0, it was established that the lattice is found in a phase where $S_A = -S_B \sim 0.874560$ and $Z_A = Z_B = 0$. This can be interpreted as a strong preference for the lattice to wrap on octahedra in the FCC lattice. In this phase, clearly the full average value $S \equiv (S_A + S_B)/2 = 0$ is not a good quantity and the correct order parameter is $S_{st} \equiv (S_A - S_B)/2$, or more simply S_A itself.

We now come to the question of the precise form of the probability distribution for the quenched disorder variables τ and w. As has been discussed previously, these disorder variables should obey the two folding constraints in order to correspond to some prior folding of the lattice. As in our previous work in [12], we can take advantage of the solution of the pure system and simply assume that the disorder distribution is described by a particular equilibrium distribution of this pure system. Since there is no physical reason to introduce an energy scale in the distribution of the disorder variables and because we want to treat as equiprobable all prefolded configurations, the natural choice is to take the distribution of the pure system at K = 0. As we just mentioned, the triangular lattice is then in a 'octahedrally' folded phase. The disorder configuration will thus also have the same nature, i.e. $\overline{\tau_{st}} = 0.874560$. This means that the disorder configuration is dominated by obtuse and complete creases. We will refer to the disordered model with this particular probability distribution as model 1. For comparison, we will also study a model without the physical constraints on the τ and w variables, i.e a case were the two random variables take ± 1 values with equal probability, independently on each triangle. All the $(2 \times 2)^6 = 4096$ hexagonal disorder configurations are then possible and equiprobable. We shall refer to this second model as model 2 and will compare its behaviour with that of model 1.

To analyse the properties of both models 1 and 2, we rely on the same method we used in [12] for the planar case, i.e. the cluster variation method (CVM) in its hexagonal approximation. The CVM uses a variational principle on the free energy of the system together with a suitable truncation of the cumulant expansion of the entropy at the level of some maximal clusters (here the hexagons) [14–16]. It is applicable to the statistics of both pure systems without disorder and to systems with quenched random disorder. In this case, it allows one in particular to evaluate average values such as (9). This method has

been first used for the planar and FCC folding problems in [17]. Although it is only an approximation since the entropy is evaluated from a truncation of its cumulant expansion, it has been shown in the most simple cases of folding problems that its results compare extremely well with exact predictions [17]. For more complex cases where no exact results exist, we still believe that this method allows for reliable and accurate predictions, as far as the nature of the phases and of the transitions are concerned. We refer to our previous work [12] for a detailed description of this method and its implementation for disordered systems.

We now present the results of our CVM analysis. Our main result is, as for the planar system, the dependence of the existence of a frozen phase $F_i \neq 0$ (i = 1, 2) on the type of disorder. In the case of model 1, a discontinuous transition occurs at $K = K_{F3} \sim 0.44$ from a low K octahedrally folded phase to a large K frozen phase. Model 2 does not develop such a phase and the lattice is always found in the octahedrally folded state (this was checked up to K = 10.0). We have also looked at the effect of the external field H_r alone. In this case, both systems show a discontinuous transition to a partially frozen phase $0 < F_i < 1$ with, however, very different resulting states. In the frozen phase of model 1, the F_i almost saturate to 1 and the lattice is thus completely trapped in the configuration specified by the disorder variables. In the case of model 2, the values of F_i are much smaller, which means that the degree of freezing is far from complete. Such a different character comes from the frustration appearing in the system for model 2, a result similar to what we found in the planar folding model [12]. The external field tries to put the lattice configuration into a disorder configuration which is, in general, not accessible due to the folding constraints on the σ and z variables. In other words, there is not a unique ground state for arbitrary disorder variables.

In figures 1 and 2, we show the behaviour of the free energy and the different order parameters S_A , Z_A , $F_{1,C} \equiv F_1 - S_A \overline{\tau_A}$ and F_2 for both model 1 and model 2 as a function of the bending energy K and the external field H_r . We display $F_{1,C}$ instead of F_1 , because in the case of model 1, F_1 takes non-zero values even at $(K, H_r) = (0, 0)$. Indeed, when $K = H_r = 0$, the disorder variables τ and w are decoupled from the physical degrees of freedom σ and z. From the definition of F_1 , its value is then simply given by the product of the value S_A of the pure system at K = 0 and the quantity $\overline{\tau_A}$, both equal to 0.874560, leading to $F_1 = (0.874560)^2 \sim 0.765$. This non-zero value is subtracted if we use the 'connected part' $F_{1,C}$ instead.

In the case of model 1, by increasing the bending rigidity K, the systems undergoes a phase transition from the octahedrally folded phase ($S_A \neq 0$, $Z_A = 0$, $F_{1,C} \neq 0$ and $F_2 = 0$) to the frozen phase ($S_A \sim \overline{\tau_A} = 0.874560$, $Z_A = 0$, $F_{1,C} \sim 1.0 - (0.874560)^2 \sim 0.235$, $F_2 \sim 1$). The values of F_i saturate to 1, meaning that the system is almost completely trapped in the configuration specified by the disorder variables. The external field also induces a phase transition. In this case, a weak field already creates a non-zero value of F_2 and the lattice is thus in a weakly frozen phase. A discontinuous transition occurs to a highly frozen phase at with strong freezing $F_i \sim 1$ (i = 1, 2).

In the case of model 2, increasing the bending rigidity does not give rise to a frozen phase (in the range K < 10.0 at least where we performed our analysis). The octahedral order $S_A \neq 0$ persists for all K and the triangular lattice remains in the octahedrally folded phase. With the external field H_r , the octahedral order disappears abruptly and a partially frozen phase $F_i \neq 0$ (i = 1, 2) appears at $H_{r,C} = 0.18$. However, the values of F_1 and F_2 are small in this phase compared with the large H_r frozen phase of model 1.

In the present work and in our previous work [12], we have studied triangular lattice folding models with random bending rigidity and random spontaneous curvature. These

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Figure 1. The free energy and the order parameters S_A (broken), Z_A (full), $F_{1,C}$ (broken) and F_2 (full) as a function of the bending energy *K* and the external field H_r . These results are for model 1, i.e. with disorder variables satisfying physical constraints.



Figure 2. The free energy and the order parameters S_A (broken), Z_A (full), $F_{1,C} = F_1$ (broken) and F_2 (full) as a function of the bending energy K and the external field H_r . These results are for model 2, i.e. with disorder variables completely random.

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models can be seen as toy models for crumpled paper in the sense that they combine the effect of disorder and of geometrical metric constraints on a geometrical two-dimensional object. We have in mind the situation where the paper is crumpled only once and the system thus does not have frustration and can recover this crumpled state. A natural question is now what happens if the paper is crumpled several times. In this case, we expect that the lattice should store several creases configurations. Let us discuss now how this can be implemented in our model. For simplicity we return to the *planar* folding problem with random bending rigidity (see equation (1)). We can imagine that the lattice, after *p* crumpling processes, stores *p* different configurations, which we denote by $\tau_i^{\mu}(\mu = 1, ..., p)$. Then one possible choice is the following bending energy, written by analogy with the Hopfield model [18, 19]:

$$K_{ij} = \frac{K}{p} \sum_{\mu=1}^{p} \tau_i^{\mu} \tau_j^{\mu}.$$
 (10)

We note, however, two differences with the usual Hopfield model. In our problem, the spin variables σ_i and the disorder random variables τ_i^{μ} correspond to folded configurations of the lattice, and must satisfy the planar folding constraint (equation (2)). Moreover, in the Hopfield model, the interaction is long-ranged $(\sum_{(i,j)})$, while in our model, it is simply short-ranged $(\sum_{n.n.(ij)})$.

The choice (10) for the bending energy is, however, somewhat unsatisfactory. To see why, we consider an elementary hexagon which tries to store the two disorder configurations of figure 3. With the definition of equation (10), the bending rigidity K_{ij} at the creases becomes zero (see figure 3(a)), a result which is somewhat unrealistic. We would instead expect that, if one folds the hexagon as in figure 3, the bending rigidity becomes negative at all the creases and remains positive where there is no crease (see figure 3(b)). A more natural choice for the bending energy is then

$$K_{ij} = K \min[\tau_i^{\mu} \tau_i^{\mu} (\mu = 1, \dots, p)].$$
(11)

This choice corresponds to a complete irreversibility of the process of marking creases.



Figure 3. An elementary hexagon with two different memories τ^1 and τ^2 . We show on the righthand side the corresponding values of K_{ij} according to (a) equation (10) and (b) equation (11).

With such a disorder, we now find for the case of figure 3 seven ground states in competition (figure 4). This situation is exactly similar to the case of unphysical disorder, where the τ variables do not satisfy the folding constraint. In our previous work, we have shown that the strong frustration caused by such an unphysical disorder prevents the system from having a frozen phase at large *K*. Based on this result, we could conclude that, in the presence of many different stored configurations, the system remains always disordered. On the other hand, as we increase the number of irreversible crumplings, the number of creases



Figure 4. An elementary hexagon with unphysical disorder and the corresponding lowest energy configurations satisfying the folding constraint.

becomes large and the system becomes almost identical to a *pure* model with negative bending rigidity. Such a pure system is known to be at large K in a so-called 'piled-up' ordered phase. We can assume that one develops such an order if the number p of stored configurations exceeds a critical value p_{c2} . Finally, from the properties of the Hopfield model, one can also imagine the system to be in the so-called spin-glass phase.

From the above discussion, we can propose the following conjecture on the statistical properties of the above model at $K = \infty$, which is of particular importance because crumpled paper can be considered as an infinite elastic constant limit $K \to \infty$ (or T = 0) of the lattice system. The triangular lattice with p stored configurations could be in the retrieval phase for very small $p < p_{c1}$, where the system can almost store and retrieve p patterns. For $p_{c1} , the system would be in a spin-glass phase or in a disordered phase. Finally, for <math>p > p_{c2}$, the system would be in the piled-up phase. The values of p_{c1} and p_{c2} are expected to be very small, because the interaction is short ranged and the probability that each elementary hexagon has two or three creases is very high, even with one stored configuration.

Finally, we also expect that the many crumpled configurations which are caused by successive crumplings of the paper are not independent from one another and we can suppose that they look quite similar. This weakens the frustration, which might keep the paper in the retrieval phase for larger p, i.e. increase the value of p_{c1} and p_{c2} . In fact, it was also reported experimentally [5] that the paper has a good memory of the previous crumpled configuration. Even in the retrieval phase, the energy landscape in the phase space of the system can be very complex. It is this complex nature which might appear as the universal power law of the noise emitted from the crumpled elastic sheets [5]. The thermodynamics of the above model are left for a future study. In addition, more realistic bending energy could be considered. To conclude, we can say that the problem of multiple stored configurations is still very open.

We thank Dr M Bowick and Dr Y Ozeki for useful discussions.

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