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Long range topological correlations in cellular patterns

C. Oguey *

LPTM, CNRS UMR 8089, University of Cergy-Pontoise, 2 ave A. Chauvin, 95302 Cergy-Pontoise, France

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ABSTRACT

In random 2D cellular assemblies, maximum entropy inference yields a specific form for the topological pair correlation, bi-affine in the cell charges (6-polygonality). By a self-consistent method, we show that the long distance behaviour of the pair correlation function is related to sum rules involving moments of the quadratic coefficients of the bi-affine form. The correlation function is predicted to decay like the 4th power of inverse distance if the first moment does not vanish, faster otherwise. The lowest sum rule expresses the screening of topological charge in the foam. Comparison with sparse available experimental data is not conclusive.

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1. Introduction

Foams are random but finding their probability distribution is still an intriguing challenge. Even if all observables are prone to follow some distribution laws, the present study is devoted to elementary topological aspects such as counting cell sides, neighbourhood, stepwise distance, with main focus on two-body correlations. Space-filling and randomness are essential, common characteristics of foams; the question addressed here is what are the implications of those properties alone in the structure and rigidity of foams, independently of the geometrical and energetic details specific to each sample of the tremendous variety of physico-chemical systems forming cellular aggregates. So far, only a few investigations have been devoted to correlations beyond nearest neighbours. Pair correlations at arbitrary distance were analysed in [1,2,3,4,5], and measured in [4].

The inevitable constraint of space-filling conditions most of the topology. Disorder is treated by Maximum Entropy, one of the few methods able to predict some aspects of the probability distributions [6]. Maximum entropy (maxent) arguments yield a specific form for the pair correlations at arbitrary distance [2].

The present analysis is devoted to bi-dimensional foams, subject to extensive theoretical and experimental investigations [1–4,6–25].

In [5], we showed that the asymptotic behaviour of layer populations was controlled by sum rules for the maxent coefficients of the pair correlator. But a closer examination of the asymptotic conditions, in a self consistent way, shows that these sum rules also constrain the decay of the pair correlation: either as j^{-4} if the first moment $M_1 \neq 0$, or faster if $M_1 = 0$, as a function of distance j.

2. Foam statistics

For consistency, we briefly recall some elements of foam topology and statistics. Details can be found in [5].

A foam $\mathcal{F}$ divides space into $N = |\mathcal{F}|$ polygonal cells. Here $\mathcal{F}$ is viewed as a set of cells and $|\mathcal{F}|$ represents the number of elements in the set.

2.1. One cell statistics

The local observable is n, the number of sides of each cell (“polygonality”), or the *topological charge* $q = 6 - n$. The fraction of $(6 - q)$-sided cells is $p(q) = N(q)/N$.

* Tel.: +33134257518.
E-mail address: oguey@ptm.u-cergy.fr

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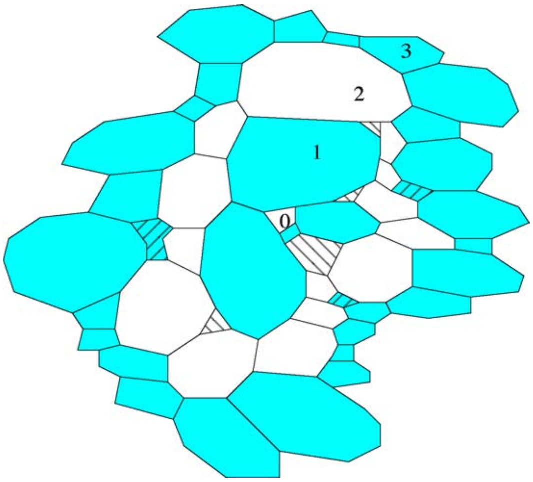

Fig. 1. Stratification into layers around a cell 0. The defects are marked hatched.

The laws of Plateau, vertex coordination (or degree) $z = 3$, and Euler imply

$$\langle q\rangle = \langle 6-n\rangle = \sum_{q<6} q p(q) \rightarrow 0 \text{ as } N \rightarrow \infty. \tag{1}$$

under mild assumptions on the foam boundary.

The second moment is $\mu_2 = \langle q^2 \rangle = \langle (n-6)^2 \rangle$.

2.2. Pair of cells, correlations

The topological distance j between two cells is the minimal number of steps needed to join the cells, where a step is a move from a cell to a contiguous neighbour cell. For example, in Fig. 1, cells 0 and 2 form a $(2, -5)$ pair at distance $j = 2$. The j th *layer* around a given cell o, lay $(j|o)$, is the set of cells at distance j from o (Fig. 1). It has population $K_j(o) = |\text{lay}(j|o)|$. The average over $(6-q)$-sided central cells is $\langle K_j(q)\rangle$ and the overall average is $\langle\langle K_j \rangle\rangle$.

The joint distribution $p_j^{(2)}(q_1, q_2)$ — probability that a (q_1, q_2) -pair of cells occurs at mutual distance j – and the corresponding marginal distribution $s_j(q) = \sum_{q'} p_j^{(2)}(q, q')$ — probability that a cell is at distance j from a $(6-q)$ -sided one — satisfy $s_j(q) = \frac{\langle K_j(q)\rangle}{\langle\langle K_j\rangle\rangle} p(q)$.

The correlator $A_j(q_1, q_2)$ and correlation function $g_j(q_1, q_2)$ are defined by

$$p_j^{(2)}(q_1, q_2) = A_j(q_1, q_2)\frac{p(q_1)p(q_2)}{\langle\langle K_j\rangle\rangle} \tag{2}$$

$$p_j^{(2)}(q_1, q_2) = g_j(q_1, q_2) s_j(q_1) s_j(q_2). \tag{3}$$

Both account for the statistical dependence of the simultaneous occurrence of a (q_1, q_2) -pair of cells at distance j. They only differ by the way they are normalised: the correlation function is 1 whereas the correlator is $\langle\langle K_j \rangle\rangle$ in independent situations.

The following consistency (or tautological) identities were proved in [1,2], involving the average population and charge of layer j:

$$\sum_{q'} p(q') A_j(q', q) = \langle K_j(q)\rangle \tag{4}$$

$$\sum_{q'} q' p(q') A_j(q', q) = \langle Q(\text{lay}(j|q))\rangle \tag{5}$$

Averages are conditioned by the central cell having $(6-q)$ sides. The charge Q of a set of cells —here, a layer— is the sum of the individual charges.

Remark. The "average charge per cell" in j -layers, $\prec q_j(q) \succ \equiv \langle Q(\text{lay}(j|q))\rangle / \langle K_j(q)\rangle$, appears in the Aboav–Weaire law (for $j = 1$).

3. Recurrence equation

3.1. Recursion equation

The central equation was derived in (1) and (2):

$$\Delta\langle K_j(q)\rangle + \langle Q(\text{lay}(j|q))\rangle = \langle I_j(q)\rangle \simeq 0, \tag{6}$$

where $\Delta K_j = K_{j+1} - 2K_j + K_{j-1}$ is discrete Laplacian and $\langle I_j(q)\rangle$ is a contribution due to defects, the cells of layer j which have no edge in common with the next layer, $j + 1$ (Fig. 1). It is assumed that, on average, this contribution vanishes: $\langle I_j(n)\rangle = 0$.

3.2. Maximum entropy

Extending a bi-affine formula first introduced for nearest neighbours ($j = 1$) [26], maximum entropy arguments (maxent) and the recursion relation (6) give the following expressions [2]

$$A_j(q_1, q_2) = b_j - a_j(q_1 + q_2) + \sigma_j q_1 q_2, \tag{7}$$

$$\langle K_j(q)\rangle = b_j - a_j q, \tag{8}$$

where σ_j, a_j, b_j are real parameters for $j = 1, 2, \ldots$. In the infinite foam limit, the average of (8) gives $\langle\langle K_j \rangle\rangle = b_j$.

From (2), (3), (7) and (8), one deduces the correlation function

$$g_j(q_1, q_2) - 1 = \frac{A_j(q_1, q_2)\langle\langle K_j\rangle\rangle}{\langle K_j(q_1)\rangle\langle K_j(q_2)\rangle} - 1$$
$$= \frac{\dfrac{\sigma_j}{b_j} - \left(\dfrac{a_j}{b_j}\right)^2}{(1 - \dfrac{a_j}{b_j} q_1)(1 - \dfrac{a_j}{b_j} q_2)} q_1 q_2. \tag{9}$$

3.3. Asymptotic freedom

In normal systems of statistical physics, distant events become uncorrelated. In foams, this was first measured by [4].

$$\text{As } j \rightarrow \infty,\ g_j(q_1, q_2) - 1 \rightarrow 0. \tag{10}$$

In terms of the maxent coefficients (7), (8), using (9), the decay of $g_j - 1$ amounts to

$$\frac{\sigma_j}{b_j} - \left(\frac{a_j}{b_j}\right)^2 \rightarrow 0. \tag{11}$$

In fact, following [5], we assume slightly stronger conditions:

$$\frac{a_j}{b_j} \rightarrow 0, \tag{12}$$

$$|\sigma_j| = O(j^{-2-\alpha}) \quad \text{for some } \alpha > 0. \tag{13}$$

These conditions imply (11) because $b_j \gtrsim j$ as $j \rightarrow \infty$ (Section 4.1).

The limit (12) expresses the fact that $\langle K_j(q)\rangle \rightarrow \langle\langle K_j \rangle\rangle$ and $s_j(q) \rightarrow p(q)$: conditioning by the central cell charge (for $\langle K_j(q)\rangle$, see Section 2.2) or presence (for $s_j(q)$) has a vanishing effect at large distance.

For (13), we will see, in Section 4.2, that $\{\sigma_j\}$ is related to the average charge excess induced by conditioning on a central charge q. So, again, (13) expresses the asymptotic decay of influence.

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4. Asymptotic behaviour and sum rules

4.1. Solutions and sum rules

With the maxent form (7), (8) of the correlator A_j and population $\langle K_j(q)\rangle$, the recursion relation (6) implies a system of recurrence equations for the coefficients a_j, b_j and σ_j [2,5]. Define $f_k = -\mu_2\sigma_k$. By assumption, $|f_k| \to 0$ fast at large k. So, $\sum_{k=1}^{j-1} f_k \to S$ as $j\to\infty$ and f/S defines a normalised distribution (not necessarily positive). Let the moments of this distribution be $M_n = \frac{1}{S}\sum_{k\geq 1} k^n f_k$. The solutions of the recursion equation with maxent ansatz are [5]:

$$a_j = \sum k f_k + j(1 - \sum f_k),$$
$$b_j = \frac{\mu_2}{6}\left[(j+1)j(j-1)(1-\sum f_k) + (3j^2-1)\sum k f_k - 3j\sum k^2 f_k + \sum k^3 f_k\right] + 6j.$$

All sums run from $k = 1$ to $j - 1$.

As $j\to\infty$, the coefficients of A_j (7) behave like polynomials in j [4,5]:

$$a_j \to j(1-S) + SM_1,$$
$$b_j \to \frac{\mu_2}{6}S\left[(j+1)j(j-1)(\frac{1}{S}-1) + 3j^2M_1 - 3jM_2 + M_3 - M_1\right] + 6j,$$

The polynomial order in j is lower if $\{f_k\}$ satisfy sum rules. Indeed, if $S = 1$ and $M_1 = 0$,

$$a_j \to 0$$
$$\langle K_j(q)\rangle \sim b_j \sim j.$$

This regime is the Euclidean scaling [3,7,27].

4.2. Topological charge

A physical interpretation of the moments is given by the average charge contained in a ball or in a layer.

The average excess charge in a topological ball B_j due to conditioning on $(6-q)$-sided central cells is

$$\langle Q(B_j(q))\rangle - \langle\langle Q_j\rangle\rangle = q\left[1 - \sum_1^j f_k\right] \simeq q(1-S).$$

where $\langle\langle Q_j\rangle\rangle$ is the average $\sum_q p(q)\langle Q(B_j(q))\rangle$. So $S = 1$ means global neutrality (centre + cloud).

4.3. Layer charge and Aboav–Weaire law

Formulae (7), (8), and the recurrence Eq. (6) yield an expression for the "average charge" in layer j as a fractional linear function of q:

$$\prec q_j(q)\succ = \frac{\langle Q(lay(j|q))\rangle}{\langle K_j(q)\rangle} = -\frac{\mu_2 a_j + f_j q}{b_j - a_j q}. \quad (14)$$

For $j = 1$ (nearest neighbour layer), Eq. (14) specialises to

$$\langle q_1(q)\rangle = -\frac{qf_1 + \mu_2}{6-q} = f_1 - \frac{6f_1 + \mu_2}{6-q}, \quad (15)$$

which is a form of the Aboav–Weaire law [28,29].

5. Decay of correlations

From (9), the pair correlation function is

$$g_j(q_1, q_2) - 1 \simeq \left(\frac{\sigma_j}{b_j} - \left(\frac{a_j}{b_j}\right)^2\right) q_1 q_2 \quad \text{as } j\to\infty. \quad (16)$$

Now the estimates for a_j, b_j yield

$$g_j(q_1, q_2) - 1 \sim \begin{cases} j^{-4} & \text{if } M_1 \neq 0 \\ j^{-1}\sigma_j & \text{if } S = 1 \text{ and } M_1 = 0. \end{cases}$$

In the first case, $M_1 \neq 0$, the fairly slow decay $\sim j^{-4}$ does not depend on the exact value of the sum S, equal to 1 or not. In the second case, when both $S - 1 = M_1 = 0$, the decay is different, $j^{-1}\sigma_j \sim j^{-3-\alpha}$ according to our assumption (13), faster than j^{-4} if $\alpha > 1$.

6. Discussion

Under 'minimal' assumptions of maxent bi-affine form for pair correlations, no defect contribution on average and asymptotic decay of correlations, we showed that (1) the layer population is bounded by a polynomial function of the distance j; this polynomial is of order 3, or less if the bi-linear coefficients f_j satisfy sum rules; e.g. $S = 1$ and $M_1 = 0$ imply a population growing linearly. (2) The pair correlations decay as j^{-4} if $M_1 \neq 0$, faster otherwise.

6.1. The results

Our results are not entirely predictive because we have no a priori estimates of the moments S and M_k. We can only state the equivalence between sum rules and decay rates of the correlations. Our first sum rule, $S = 0$, means that the total induced charge is zero. Global neutrality in a closed space is a consequence of Euler's theorem, but boundary terms can be important in finite samples. We do not know whether charge neutrality holds at an intermediate scale. In electrostatics, charge neutrality results from energy minimisation, requesting the shielding the long range Coulomb field. The vanishing of higher multi-polar moments of the induced charge, leading to strong Debye screening, only occurs in the high temperature plasma phase, and may thus be seen as an entropic effect. To what extent this analogy applies to foams is still a question. An energy-based analogy with electrostatic was proposed in [23], but here we have ruled out any energy contribution from the start, as in a high temperature phase.

For comparison, as far as we know, the only experimental measurements of correlations beyond nearest neighbours were done by Szeto et al. [4]. Some general features roughly agree with the theory, for ex. $g - 1$ changes sign as the charge product q_1q_2, which is a factor in Eqs. (9) and (16). However, the marked profile for neutral cells ($q = 6 - n = 0$) disagrees with Eq. (16), which predicts $g - 1 = 0$ (no correlation). The large distance behaviour is best seen in a log-log plot (Fig. 2). Despite oscillations of period 2, the experimental curves are quite compatible with a power law decay; but the observed exponent, around 1.5, is far from our closest prediction, 4. This long range correlation seen experimentally is quite a surprise, certainly raising questions on our premises. In the experimental conditions —foam aged and in "scaling regime"— departure from maxent may be expected if energy drives the stationary regime. Whether the peculiar type of organisation implied by scaling is compatible with maxent is an unanswered question so far. On the other hand, the experiment was done on a finite sample (less than 4000 bubbles in scaling regime, according to [30]), maybe too small for reliable measurements on asymptotic decay. A larger set of fits can be accepted if error bars a taken into account (in Fig. 2). Other experimental setups, as the effect of agitation [25], would be worth testing.

6.2. The premises

The maxent prediction for the correlator is a subject of debate [29]. The maxent theory is not a proof ab initio, but a clever

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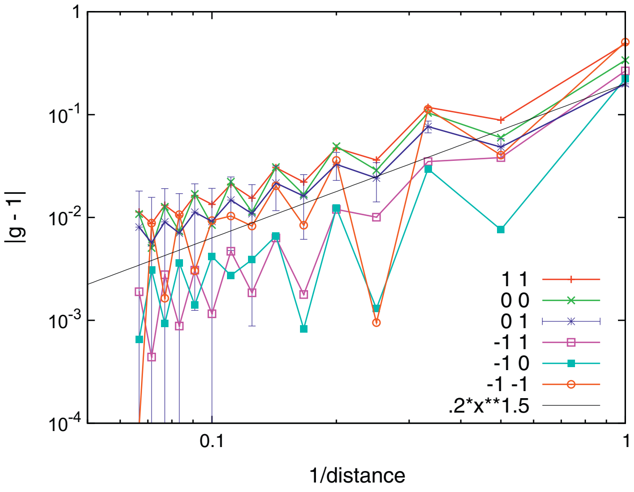

Fig. 2. Pair correlations $g-1$ vs inverse distance j^{-1} in log–log plot. Experimental data from [4]. Putative error bars are displayed only for the (0, 1) correlation.

argument based on plausible, unproven, assumptions. One of these is the freedom to adjust the correlator. Sorting out the a priori constraints and degrees of freedom in a foam is a central question [26,24]. In favour of the maxent formulae (7), (9) are the numerical simulations on variates of the “topological gas” [2,3]. On the other hand, deviations in the nearest neighbour correlations were already observed by the pioneers [26]. However, short distance examinations are not enough to refute the whole theory. The long distance asymptotics might be correct even if the short range terms deviate from reality. The first terms depend on initial conditions used to integrate the recurrence relation, or the recurrence equation itself might even prove to be incorrect for the first terms, but none of these eventualities preclude, a priori, the discrepancies to vanish at large j, as transient effects. Deriving and testing the long distance predictions directly motivated the present study.

The contribution of the defects is a big unknown in the theory, certainly requiring further research. For example, the stratification in Fig. 1 contributes by -4 to the RHS $I_2(4)$ of Eq. (6). A more detailed examination shows that, when the defect term $\langle I_j(q)\rangle$ cannot be neglected, the final correction to the populations should be negative in most cases. So our results in sec. 4.1 would remain valid as upper estimates.

I can only give a qualitative argument as to why defects don't contribute significantly, mainly based on observation [27]. In each layer, defects are present at most to a finite proportion. Layers cannot be thick; in cell steps, the individual layer thickness is $1+\varepsilon$, where $\varepsilon \geq 0$ is the contribution of defects, but the overall average thickness is 1 (layer j is at distance j from the origin and it encloses all layers $k=1, \ldots, j-1$ in between). This discrepancy comes from the fact that minimal paths, along which distance is measured, shortcut defects. The presence of defects is related to the roughness of layer profiles. Whenever curvature accumulates locally, tentatively increasing roughness about to transfer profile length to lower scales, defects appear in the depressions, fill the wells, favouring shortcuts and contributing to restore smoothness for the next layer. In brief, roughness is both necessary to and killed by the presence of defects. This balance bounds the roughness and prevents any explosion due to defects.

The a priori decay assumptions on the correlations where explained in Section 3.3. For our purposes, we assumed slightly stronger decay (12), (13) than what would be commanded by $g-1$ only. A priori, the decay assumed here was rather weak. Nonetheless, sticking to the minimal condition implied by the decay of $g-1$, Eq. (11), would be worth investigating, especially if the experimental results are confirmed.

More investigations, notably on the premises, and more experiments, are needed to lift our perplexity.

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