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A comment on Rivier's maximum entropy method of statistical crystallography

S N Chiu†

Freiberg University of Mining and Technology, TU Bergakademie Freiberg, Institut für Stochastik, D-09596 Freiberg, Germany

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Abstract. In the literature, Rivier's maximum entropy method was used to 'prove' Lewis's law and a linear Aboav's law. In this paper we show that the functional forms of these two laws for a statistically equilibrated cellular network, even if such a network really exists, cannot be derived or proved by this method. For example, within the maximum entropy method, we demonstrate that a quadratic Aboav's or Lewis's law is as probable as a linear one.

1. Introduction

Space-filling cellular structures can be observed in many natural phenomena, such as in biological tissues and in metallurgical aggregates. In the physical literature for these cellular networks it is usually assumed (implicitly or explicitly) that they consist of convex cells only and have trivalent vertices only. That means, all vertices have the same coordination number 3, since any vertex with higher coordination is structurally unstable and will split into several trivalent vertices by some small deformations (see the discussions in Delannay *et al* (1992), Le Caër (1991), Le Caër and Delannay (1993b), and Rivier (1985, 1993)). Such a network, or a random tessellation, is called *ordinary equilibrium state*. Reviews of this subject can be found in Biarez and Gourvès (1989), Bideau and Dodds (1991), Bideau and Hansen (1993), Chiu (1994b), Dormer (1980), Getis and Boots (1978), Gibson and Ashby (1988), Gorden (1978), Guinier (1980), Okabe *et al* (1992), Smoljaninov (1980), Stoyan *et al* (1987) Stoyan and Stoyan (1992), Thompson (1917) and Weaire and Rivier (1984).

Rivier (1985, 1986, 1990, 1991, 1993, 1994) and Rivier and Lissowski (1982) developed the maximum entropy method to 'derive' the *structural equations of state* (e.g. Aboav's and Lewis's laws stated below) for a cellular network which is in statistical equilibrium. It has been believed that the notion of entropy defines a kind of measure of randomness; *probability distribution of higher entropy* are 'more disordered', 'more random' and 'more probable' and they 'assume less' and are 'more natural' according to the interpretation in Shannon's (1948) information theory context. The general principle of the maximum entropy method is that when we make inferences based on incomplete information, we should draw them from that probability distribution which maximizes the entropy under the information, or constraints, we have.

Rivier's maximum entropy theory of statistical crystallography is based on two observations: (i) random cellular structures are usually indistinguishable, apart from the

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scale of measurement, and (ii) an 'ideal' random cellular structure is determined solely by some inescapable mathematical constraints and is in statistical equilibrium; this structure is 'the most probable' and 'arbitrariness invariant'. These two propositions led to the use of the maximum entropy method. This maximum entropy method was used to 'prove' Lewis's law by Rivier and Lissowski (1982) and Aboav's law (in some sense) by Peshkin *et al* (1991) (referred to as PSR hereinafter). Roughly speaking, Aboav's and Lewis's laws state that the mean number of edges of the neighbours and the mean area of a *typical* n-edged cell are linear functions of n; the term 'typical cell' means a randomly chosen cell in the network, where all cells are equally weighted, and for simplicity, a typical n-edged cell is called a 'typical n-cell'.

In this paper, we are going to see that this maximum entropy method cannot be used to derive the functional forms of the structural equations of state. There are infinitely many possibilities of the functional forms which maximize the entropy S to the same value, where S is defined in Rivier (1993) by

$$S \equiv -\sum_n p_n \ln \frac{p_n}{q_n} \tag{1}$$

where p_n is the probability that the typical cell of that cellular network has n edges, and q_n is a *prior probability*, or a prior of p_n. Therefore, the equivalence between the statistical equilibrium and the maximum value of this entropy is in doubt.

2. A review of Lewis's law and the maximum entropy method

Lewis (1928, 1930, 1931, 1943, 1944) observed in several planar cellular networks, at various stages of their development, a specific relationship between the average area of a typical n-cell, A_n say, and n:

$$A_n = \frac{1}{4\lambda}(n-2) \tag{2}$$

where λ is the mean number of cell-centroids per unit area.

Rivier (1991, 1993) and Rivier and Lissowski (1982) showed that *if there exists a linear relationship between A_n and n, for $n \geqslant 2$*, then Lewis's law can be obtained as follows. Let $A_n = a_0 + a_1 n$ be true, where a_0 and a_1 are some constants. As the mean area of a typical cell is always the reciprocal of the intensity of cells

$$\sum_n A_n p_n = \frac{1}{\lambda} \tag{3}$$

and the mean number of edges is 6, provided that all vertices are trivalent:

$$\sum_n n p_n = 6 \tag{4}$$

these two equations lead to $a_0 + 6a_1 = 1/\lambda$. By writing $a_1 = \beta/\lambda$, where β is some constant, we obtain

$$A_n = \frac{\beta}{\lambda}\left(n - \left(6 - \frac{1}{\beta}\right)\right). \tag{5}$$

Since a convex cell must have at least 3 edges, it is $A_2 = 0$, which yields $\beta = 1/4$. Hence Lewis's law (2) is obtained.

Then the maximum entropy method was used to show that the linear relationship between A_n and n maximizes the entropy S under constraints (3), (4) and

$$\sum_n p_n = 1. \tag{6}$$

The argument used in Rivier's maximum entropy method is that *if a constraint is made redundant, the entropy can be increased further*. Thus, if (3) is a linear combination of (4) and (6), then S is maximized subject to these two inescapable constraints only. This implies $A_n = a_0 + a_1 n$ with $a_0 + 6a_1 = 1/\lambda$ is 'more probable'.

Note that since $\{q_n\}$, the prior probability distribution, is not known, we cannot derive explicitly the $\{p_n\}$ such that the entropy S is maximized. Rivier and Lissowski (1982) originally assumed that typical cells of n edges are equally probable for all $n \geqslant 3$. Under this assumption, maximizing S is equivalent to maximizing the Shannon entropy $H \equiv -\sum_n p_n \ln p_n$.

Let $\{p_n^{\mathrm{ME}}\}$ denote the distribution such that the entropy S with an arbitrary but fixed prior $\{q_n\}$ is maximized under constraints (4) and (6) only, and the corresponding maximum entropy value under these two constraints is S^{ME}. Using the Shannon entropy H as in Rivier and Lissowski (1982), one can obtain that $p_n^{\mathrm{ME}} = (1/4)(3/4)^{n-3}$ and $S^{\mathrm{ME}} = 4\ln 4 - 3\ln 3$. If $A_n = a_0 + a_1 n$ with $a_0 + 6a_1 = 1/\lambda$, then the Shannon entropy can really be maximized to $4\ln 4 - 3\ln 3$. However, it is easy to obtain the same maximum entropy with, e.g., $A_n = a_0 + a_1 n + a_2 n^2$ where $a_0 + 6a_1 + 48a_2 = 1/\lambda$. Therefore, with respect to the Shannon entropy a quadratic Lewis's law is as probable as a linear one.

3. A review of Aboav's law

Aboav (1970) observed empirically that in cellular networks of trivalent vertices the total number of edges of cells neighbouring the typical n-cell of a planar cellular network is linear in n. This leads to the formula

$$nm_n = a_0 + a_1 n$$

where m_n is the mean number of edges of a randomly chosen neighbour of the typical n-cell, and a_0 and a_1 are constant such that $a_0 + 6a_1 = 36 + \mu_2$ with $\mu_2 \equiv \sum_n (n-6)^2 p_n$. The relation of the constants is due to the so-called Weaire's (1974) sum rule. However, such a linear Aboav's law is not universally true, as can be seen from the following exact identity. A rigorous proof can be found in Chiu (1994c).

Theorem. For $p_n > 0$

$$m_n = 5 + \frac{6}{n} + \frac{\operatorname{cov}(k(n, N), N)}{np_n} \tag{7}$$

where N is the random number of the edges of the typical cell, with probability distribution $\{p_n\}$, and $k(n, l)$ is the mean number of n-edged cells which belong to the complex of the typical l-cell, when $p_l > 0$, and zero otherwise. The complex of the typical cell is the union of the typical cell and all its neighbouring cells.

Proof. This $k(n, l)$ is connected to $M_n(l)$, the mean number of n-edged cells of the typical l-cell by the following simple relation:

$$k(n, l) = M_n(l) + \delta_{nl} \tag{8}$$

where δ_{nl} equals to 1 when $n = l$ and zero otherwise.

Counting the total number of edges of all n-edged cells from the number of their adjacent cells ($\sum_l \{M_n(l) \times \#l\text{-edged cells})\}$) and from the number of their own edges

($n \times$ #n-edged cells), and then dividing the total number of cells yields

$$\sum_l M_n(l)p_l = np_n \tag{9}$$

where p_n is assumed loosely to be the relative frequency of n-edged cells. Moreover, the total number of edges between n-edged and l-edged cells ($M_n(l) \times$ #l-edged cells) is the same as that between l-edged and n-edged cells ($M_l(n) \times$ #n-edged cells). Dividing this number by the total number of cells yields

$$p_l M_n(l) = p_n M_l(n). \tag{10}$$

Note that by the same argument or simply by substituting (8) into (9) and (10), similar identities are obtained for $k(n, l)$

$$\sum_l k(n,l)p_l = (n+1)p_n \tag{11}$$

$$p_l k(n,l) = p_n k(l,n). \tag{12}$$

A few steps of calculation lead to

$$nm_n = \sum_l lk(n,l)\frac{p_l}{p_n} - n$$

$$\sum_l lk(n,l)p_l = 6(n+1)p_n + \operatorname{cov}(k(n,N),N).$$

These two equations yield (8). □

4. A maximum entropy prediction of Aboav's law

PSR used the maximum entropy method to show that in order to maximize the entropy S, $M_l(n)$ should be in the following linear form:

$$M_l(n) = A_l + B_l n \tag{13}$$

for some A_l and B_l depending on l only. This leads to a linear Aboav's law:

$$nm_n = \sum_l lM_l(n) = \left(\sum_l lA_l\right) + \left(\sum_l lB_l\right)n. \tag{14}$$

There are several constraints imposed on $\{p_n\}$, namely, (4), (6), (9) and (10) stated in the previous sections. If $M_l(n)$ is written in the linear form (13), then (9) can be re-expressed as a combination of (4) and (6), and so the entropy S can be increased further.

Moreover, (9) is a consequence of (10), which is made redundant by the linear form (13); this has not been established in their paper.

To show this redundancy of (10), note that substituting (13) and (8) into the exact identity (7) yields

$$nm_n = 6n + \frac{B_n}{p_n}\mu_2 \tag{15}$$

as $\operatorname{cov}(\delta_{nN}, N) = (n-6)p_n$. Compare (15) with (14) we obtain

$$\frac{B_n}{p_n}\mu_2 = \left(\sum_j jA_j\right) + \left(\sum_j jB_j\right)n - 6n, \tag{16}$$

for reasons which will become clear, the running index is changed from l to j.

By (4), (6) and (9), (13) becomes

$$A_j + 6B_j = jp_j. \tag{17}$$

Multiplying both sides of (17) by j and then summing up all possible j yields

$$\sum_j jA_j + 6\sum_j jB_j = \mu_2 + 36. \tag{18}$$

Substituting (18) into (16), yields

$$\frac{B_n}{p_n}\mu_2 = \mu_2 + (n-6)\left(\sum_j jB_j\right) - 6(n-6). \tag{19}$$

Therefore, from (13), (17) and (19),

$$\frac{M_n(l)}{p_n} = n + l - 6 + \frac{(l-6)(n-6)}{\mu_2}\left(\sum_j jB_j - 6\mu_2\right) = \frac{M_l(n)}{p_l}.$$

Thus (10) is fulfilled.

5. Other entropic predictions of Aboav's law

Let us reconsider PSR's argument again. If constraints (11) and (12) instead of (9) and (10) were used in their argument, they would conclude that $k(n, l)$ is linear in n, too. That is

$$k(l, n) = A'_l + b'_l n \tag{20}$$

where A'_l and B'_l are some constants depending only on l. Although a linear Aboav's law still results, it is impossible to have both $M_l(n)$ and $k(l, n)$ being linear in n, since they are different by 1 at the point $l = n$ and are the same otherwise. However, the entropy S in this case is still S^{ME}, since with the linear form (20), it can be easily shown that

$$\frac{k(n, l)}{p_n} = n + l - 5 + \frac{(l-6)(n-6)}{\mu_2}\left(\sum_j B'_j - \frac{6}{\mu_2}\right) = \frac{k(l, n)}{p_l}$$

and so (12), as well as (9), (10) and (11), is also redundant. Therefore, no matter if it is $M_l(n)$ or $k(l, n)$ (but not both) that is linear in n, the entropy is still S^{ME}, and so it is not clear which functional form is 'more probable'.

It is also possible to establish another entropic prediction of the form of $M_l(n)$ with the same entropy value S^{ME}, by making (9) to be a linear combination of (4) and (6). Define

$$f(l, n) = M_l(n) - \delta_{nl} l = k(l, n) - \delta_{nl}(l + 1).$$

Then both constraints (9) and (11) can be rewritten as

$$\sum_n f(l, n) p_n = 0. \tag{21}$$

Using the argument of this maximum entropy method, the entropy S can be increased further when $f(l, n)$ is in the form

$$f(l, n) = a_l + b_l n \tag{22}$$

for some constants a_l and b_l depending on l only, provided that $a_l + 6b_l = 0$. This implies $M_l(n)$, $k(l, n)$ and nm_n are nonlinear:

$$nm_n = \sum_l lM_l(n) = n^2 + \left(\sum_l lb_l\right)n - 6\sum_l lb_l. \tag{23}$$

To prove the entropy is still S^{ME}, it suffices to show that under (4), (6) and (22), (10) is redundant, and consequently so are (9), (11) and (12). A proof can be constructed easily and is given in Chiu (1994a), where further examples of different forms of nm_n are also given.

Actually, whenever $\sum_l M_l(n) = n$ and $M_l(n)/p_l$ is a function which is symmetric in n and l, (10) holds, and so does (9). Thus, no matter whether $M_l(n)/p_l$ is $(nl)/6$, $1 + (n-1)(l-1)/5$, $2 + (n-2)(l-2)/4, \ldots$, or $n\delta_{nl}/p_n$, $1 + (n-1)\delta_{ln}/p_n, \ldots$ etc, constraints (9) and (10) are redundant. Hence, the entropy can still be maximized to S^{ME}; that is to say, all these forms are 'equally probable'. That means, $M_l(n)$ is not uniquely determined by this maximum entropy method, nor is $k(l, n)$.

These examples show that it is not yet clear that the functional form(s) of which characteristic(s), $M_l(n)$, $k(l, n)$, $f(l, n)$, $M_l(n)/p_l$ or some others, can be chosen arbitrarily. Even if we can set up some criteria such that only the functional form of $M_l(n)$, say, can be chosen arbitrarily, it is not necessary to be linear. Even if it had to be linear, the slope and the intercept would not be fixed. Thus, the structure can undergo changes without changing the value of the entropy, and so the structure is not in statistical equilibrium.

6. A general maximum entropy method

Rivier's maximum entropy method can be written in the following generalized form. Consider a characteristic of a typical n-cell, $h(n)$ say, and suppose that there are only three constraints imposed on $\{p_n\}$, namely, (4), (6) and

$$\sum_n h(n) p_n = K \tag{24}$$

where K is some constant. The maximum entropy method of Rivier states that *if* the functional form of h can be chosen arbitrarily (it should be emphasized once more that so far there has been no criterion established for the *arbitrariness* of the functional form of some characteristic, but here we just suppose the functional form of h, no matter what it is, can be chosen arbitrarily), then when

$$h(n) = a_0 + a_1 n \tag{25}$$

constraint (24) is a linear combination of (4) and (6) and so is made redundant. Consequently, the entropy S can be increased further, and the structure with (25) is 'more probable'. If there are more than three constraints, the argument is essentially the same, i.e. choosing a functional form of h arbitrarily such that it is consistent with all constraints and at the same time makes some constraint(s) redundant. Then this functional form of h is 'more probable'.

A linear Lewis's law can be 'derived' by letting $h(n) = A_n$ and $K = 1/\lambda$. In the present paper, a quadratic Aboav's law (23) is obtained by using $h(n) = M_l(n) + \delta_{nl} n$ and $K = 0$; there is another constraint, but it is shown to be redundant if the linearity of h holds. However, this quadratic Aboav's law is shown to be of the same entropy value S^{ME} as that of the linear law derived in PSR. Which functional form of Aboav's law is 'more probable' is still not clear. The same problem arises in Lewis's law.

The reason of the existence of more than one equally probable functional form of h is that (25) is only a *sufficient but not necessary condition* for the entropy S to be maximized.

7. Infinitely many possibilities of the functional form of $h(n)$

Assume that there are only three constraints (4), (6) and (24) imposed on $\{p_n\}$. We do this since no matter how many constraints there are, the argument is essentially the same. The aim of Rivier's maximum entropy method is to make (24) redundant by using (25) with $a_0 + 6a_1 = K$. However, suppose that we first maximize the entropy subjected to (4) and (6). The solution is p_n^{ME} with the entropy S^{ME}. Then we have a great degree of freedom to choose the functional form of $h(n)$ such that (24) holds in order to maintain the entropy at the same value.

More generally, consider an arbitrary sequence of real-valued functions $\{g_i\}$ which are defined on all integers. Let

$$\sum_n g_i(n) p_n^{\mathrm{ME}} = G_i \tag{26}$$

provided that G_i's exist and are bounded. Let

$$h(n) = \sum_i a_i g_i(n) \tag{27}$$

$$\sum_i a_i G_i = K. \tag{28}$$

Constraint (24) is still satisfied by $\{p_n^{\mathrm{ME}}\}$ and so is redundant. This implies that the entropy is still S^{ME}. However, the sequence $\{g_i\}$ in (27) cannot be 'derived' by the maximum entropy method. All g_i's can still be chosen arbitrarily without reducing the entropy, provided that the corresponding a_i's in (27) satisfy (28). Thus, there are infinitely many possible functional forms of h such that the entropy is always maximized to be S^{ME}.

Note that (26) and (28) are not imposing new constraints on $\{p_n\}$, but on $\{a_i\}$, i.e. when a functional form is given, the condition (28) imposes a constraint on the coefficients of the functional form in order to maximize the entropy.

Therefore, *the maximum entropy is always S^{ME}, no matter how (24) is made redundant.* (A redundant constraint is a constraint that $\{p_n^{\mathrm{ME}}\}$ and S^{ME} are the same with or without this constraint. Another possible definition is that the solution space is the same with or without this constraint. Obviously, the former definition is more suitable than the latter in this context. Otherwise, we have to say 'to make a constraint do not discard the solution which is obtained by ignoring this constraint'.)

8. Conclusion and Discussion

It is shown by examples that Rivier's maximum entropy method does not lead to a unique specific functional form of a structural equation of state of a statistically equilibrated cellular network, even if such a network really exists. The equivalence between the statistical equilibrium and maximum value of the entropy S is not yet established. Suppose S is maximized by some functional form of a structural equation, the statistical structure of the network can still undergo some changes without reducing the entropy. Therefore, it is more likely that they are not equivalent, otherwise the statistical equilibrium may not exist.

The fallacy of this method comes from the confusion between the *sufficiency* and *necessity*. This method assumed that the statistical equilibrium leads to the maximum entropy and then showed that a linear Aboav's or Lewis's law implies that the entropy is maximized. That is,

a linear Aboav's law $\Longrightarrow$ S is maximized $\Longleftarrow$ statistical equilibrium.

However, when the entropy S is really maximized, it is not clear whether or not a unique linear Aboav's law holds and whether or not the froth is in statistical equilibrium. The examples presented above show that the argument in PSR does *not* lead to the following conclusion:

a unique linear Aboav's law $\Longleftarrow$ S is maximized $\Longrightarrow$ statistical equilibrium.

Note that the linearity assumption is stronger than the assumption that the entropy is maximized. Thus, the equivalence between the linearity of A_n, nm_n or $M_l(n)$ and the statistical equilibrium is *not excluded* in the present paper.

One consequence of the results presented here is that this method also cannot be employed to find any hidden constraint, as we show in the following example.

Gervois *et al* (1992) and Lemaître *et al* (1992, 1993) found that in the experiments of hard discs on an air cushion table

$$A_n = a + bn + \frac{c}{n} \tag{29}$$

where a, b and c are some constants. They suggested that there is a hidden constraint on this experiment, namely, $\sum_n (1/n)p_n = Z$, for some constant Z.

However, if $a + 6b + \mu_{-1}c = 1/\lambda$, where $\mu_{-1} \equiv \sum_n (1/n)p_n^{\mathrm{ME}}$, the entropy is still maximized to S^{ME} with the functional form (29) but without the extra constraint $\sum_n (1/n)p_n = Z$.

Note that the argument here is valid generally even if there are some other constraints, except that the constraints imposed on h and $\{p_n\}$ are strong enough to guarantee that h in (24) is unique, and so h can be found by solving the usual maximization problem. Then the functional form of h is no longer arbitrary, and it is not possible to increase the entropy further by making some constraint or constraints redundant. However, whenever it is possible to take an arbitrary form of h, neither the maximum entropy method can be employed to derive a unique structural equation of h, nor can it be determined that the network is statistically equilibrated.

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