Rings and Things

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Contents

List of Notes By David

List of Notes By Mark

1 | Introduction

The notion of describing amorphous materials as random networks dates back to Zachariasen, who in 1932 sketched a simple diagram of a two-dimensional glass [1]. This configuration, reproduced in figure 1.1a, showed a collection of percolating rings with an absence of long-range order. At the time, Zachariasen's image was intended only as schematic to illustrate the analogous effects in three-dimensional glasses. However, some eighty years later, modern synthesis techniques have led to a range of two-dimensional materials including amorphous carbon, silica and germania which can be considered realisations of Zachariasen's glass [2–6]. These advances may yet represent a watershed moment in chemistry, facilitating the development of a wide range of technologically useful materials with applications including catalysis and gas separation [7–9].

It is clear that understanding the structure of amorphous materials is key to this aim. However, due to the relative recentness of these experimental discoveries, much of the existing theory arises from studies of systems on greater length scales. Specifically, in the second half of the 20th century, much work was done on the formation of polycrystals in metals and alloys. By annealing the metal and slicing through the sample, the grains in the polycrystal could be directly imaged; revealing a system of tessellating polygons not dissimilar to an atomic material [10, 11]. Over time it became apparent that the structure of these networks is constrained on a series of different levels.

Firstly the mean ring size (*i.e.* the average number of sides in a polygon) tends to the constant value of six. This is readily explainable via graph theoretic arguments, simply resulting from Euler's formula when each vertex forms part of three edges - as is the case for trivalent atoms or the meeting of three grain boundaries. Intuitively

from chemistry we know this to be true: a pristine graphene sheet is a hexagonal net and although a Stone-Wales defect introduces pentagons and heptagons, they occur in pairs to preserve the overall mean ring size [12].

The next level of information is then the explicit distribution of polygon sizes, also known as the ring statistics. With the constraint of a fixed mean, the ring statistics were shown to be relatively well defined, following a log-normal or maximum entropy distribution [13–15]. However, the ring statistics alone are not sufficient to fully describe the network topology. This is because the same set of rings can be arranged in a large number of different ways. Consider again Zachariasen's original configuration. Removing one square achieves a mean ring size of six and allows the constituent rings to be arranged as a periodic tiling. Figures 1.1b-1.1d show three such examples tilings.
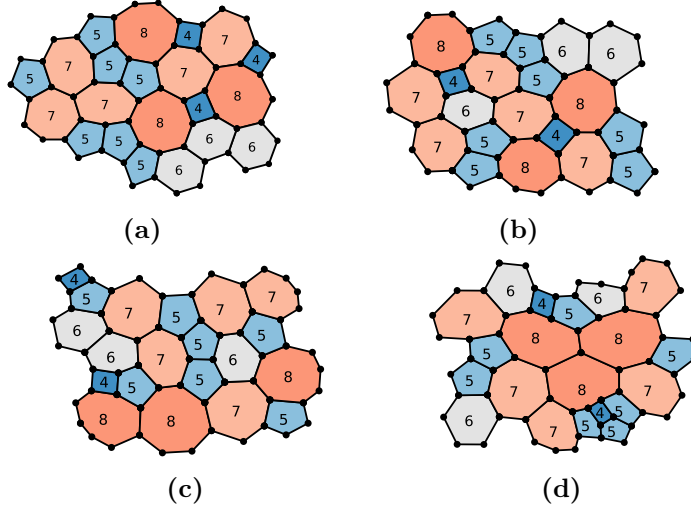

Figure 1.1: Panel (a) shows Zachariasen's glass and panels (b)-(d) three different periodic arrangements based on the glass (with one square removed to satisfy Euler's formula). Moving from panel (b)-(d) there is increased clustering of similar sized rings. The size of the rings are highlighted numerically and by colour.

Whilst they may initially look similar, on closer inspection the three configurations display fundamentally different properties. In figure 1.1b similar sized rings are dispersed throughout the arrangement whilst in 1.1d they are tightly clustered together. Furthermore, given the large number of configurations which may be theoretically possible for any set of ring statistics, only a subset of these

may be physically realisable. Empirically, these are found to be the ones in which large rings tend to be surrounded by smaller rings *i.e.* similar to 1.1b. Once again, chemical intuition would support this in the context of atomic materials, as strain is minimised by maintaining bond lengths and angles as close to their equilibrium values as possible. This effect was first noticed in polycrystals and quantified through the Aboav-Weaire law [16, 17]. This law claims that the mean ring size about any given ring can be related to the central ring size by a single fitting parameter. Hence the value of this parameter in some way describes the increased tendency of the small rings to be adjacent to large rings. The Aboav-Weaire parameter therefore provides information on the first-order ring correlations, completing the topological description of the network material.

The novelty and potential usefulness of two-dimensional materials makes them a clear candidate for computational study, in order to complement and supplement experimental endeavours. Taking the example of thin silica films, there have already been multiple complementary computational investigations including both *ab initio* methods and molecular dynamics studies using classical force fields at varying levels of theory [18–25]. In order to perform these simulations, it is necessary to have a starting atomistic configuration. This can be achieved in multiple ways. The most straightforward is to take one of the existing experimental images. These are however limited in size and number and can contain defects or areas which cannot be fully imaged. As a result, computational techniques are often preferable, but generating configurations will the required topological properties (*i.e.* correct ring statistics *and* Aboav-Weaire parameter) has proved surprisingly difficult [26, 27]. Therefore, the first part of this thesis will focus on developing methods to generate configurations of two-dimensional networks in which the topological parameters can be tuned in a controllable manner. These configurations can then be used as a seed for further computational studies, removing the reliance for experimental configurations and opening the door for high-throughput calculations which can be speculative and potentially predictive.

However, the scope of this work extends beyond materials modelling. As previously mentioned, much of the original work in this field focussed on polycrystals of metal oxides with some links to foams and Voronoi polygons [28, 29]. It is now clear that these chemical networks fit into a much wider class of two-dimensional physical networks that are ubiquitous in the natural world, emerging across all physical disciplines and length scales. Traditional examples range from the atomic level of ultra-thin materials, through colloids, foams, epithelial cells and all the way to geological rock formations [30–34]. There are however countless more occurrences, with drying blood, stratocumulus clouds, crocodile scales and geopolitical borders all being the subject of studies [35–38]. More intriguingly, although these systems are incredibly physically diverse, they still have similar structures [39]. This is because they can all be mapped onto the same generic system, which can be equivalently described as a collection of tessellating polygons or percolating rings, and hence they are governed by the same fundamental laws. Understanding the behaviours of two-dimensional networks is therefore key to a wide range of problems in frontier research, not only the directed synthesis of nano-materials but also for example the control of mitotic division [40, 41]; as well as to curiosities such as explaining the arrangement of the stones in Giant's Causeway or cracking in famous artworks [42, 43].

Furthermore, the continuing expansion and maturity of network science as a field has led to significant advances in the description and characterisation of complex networks. This has largely been driven by interest in networks in the more abstract sense of the internet, social media and neural networks [44–46]. To date, the application of these principles in the physical sciences has mostly been confined to topics such as biological signalling pathways. The second half of this thesis will therefore show how robust metrics from network science can be applied to physical two-dimensional networks to better quantify their structure and replace the need for empirical measures such as the Aboav-Weaire law. This also has the effect of tying physical two-dimensional networks into the wider field of network science, showing them to be a unique and interesting addition to the area.

As part of this process, more generic methods will be developed to construct two-dimensional networks across a range of potential models, coordination environments and topologies. This will allow a systematic study into the factors which influence the underlying network properties in two-dimensional systems. These will be compared to two further in-depth studies of network forming structures from the physical sciences. Expand colloid/procrystals bit. The first are Voronoi tessellations formed in colloidal monolayers. which can be simulated via hard particle models [47]. The second are the ring structures in so-called "procrystalline" lattices [48]. Paragraph to wrap up

1.1   Thesis Structure

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2 | Network Theory & Computational Methods

This chapter discusses the theory underpinning complex networks, covering the representation of atomic systems as networks and the relationship of the dual network to ring structure. The laws which govern the topological properties of physical networks, namely Euler's law, Lemâitre's law and the Aboav-Weaire law are also explored. Further developments linking to modern network science etc. are given in chapter Link to network theory later . In addition some of the existing computational approaches for generating realisations of two-dimensional networks are presented. A broad overview of Monte Carlo methods is given, before specific methods are covered in detail. These include the bond switching algorithm and hard particle Monte Carlo in conjunction with the Voronoi construction. Extensions to these methods and additional approaches are outlined in the relevant chapters Link to bond switching/Voronoi/mx2/procrystals later . Finally some measures are discussed...

2.1 Network Theory

The scope of what constitutes a complex network is extremely broad, covering everything from the tangible (*e.g.* computational clusters) to the more abstract (*e.g.* social interactions). Yet part of the appeal and power of network science is the ability to quantify and relate these highly disparate systems with the same underlying theory. A network is simply a collection of components termed *nodes* and the connections between them termed *links*, an example of which is given in figure 2.1. There are then two fundamental classes of network based on the

nature of the connections. Networks in which the links between nodes are mutual are termed undirected, whereas those in which the links are one-way are termed directed [49]. At the risk of dating this thesis, this is the difference between Facebook (an undirected social network of friends) and Twitter (a directed social network of followers). All the networks considered in this work are undirected and all the theory assumes this property.
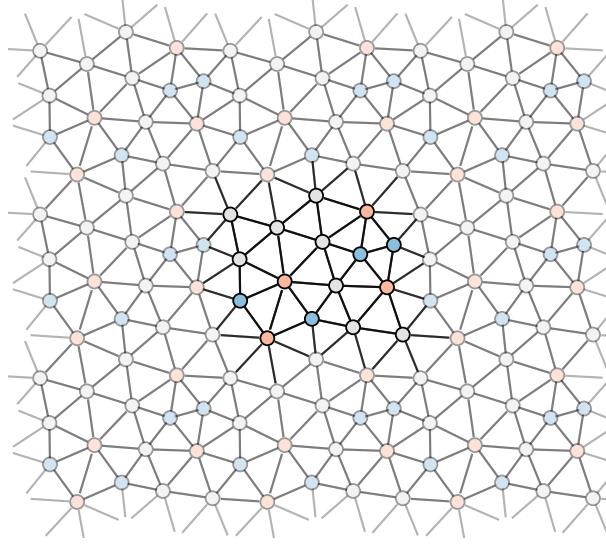

Figure 2.1: Example of a periodic two-dimensional network where nodes are represented by circles and links as lines. Nodes are coloured similarly according to their degree, whilst periodic images are greyed out to highlight the central repeating unit.

2.1.1 Node Degree and Degree Distributions

A key concept in network science is the the node degree, defined as the number of links that each node possesses. A node with k links is then said simply to have degree k, where $k \in \mathbb{N}$. This is illustrated in figure 2.1, which consists of 5- (blue), 6- (grey) and 7- (red) degree nodes. The occurrence and correlations of nodes of given degrees can then be described by a range of probability distributions.

The probability of a randomly selected node having degree k is given by the node degree distribution, denoted p_k. This is a normalised discrete distribution such that

$$\sum_k p_k = 1 \,. \tag{2.1}$$

The nth moments of this distribution are then given by:

$$\langle k^n \rangle = \sum_k k^n p_k \,. \tag{2.2}$$

Alternatively, one can also calculate the probability that a randomly selected link has a k-degree node at the end, denoted q_k. This is not the same as the distribution above, as there is greater chance of selecting links which emanate from high degree nodes, which is proportional to the node degree. As this distribution is normalised, this leads to the relations:

$$\sum_k q_k = 1 \tag{2.3}$$

$$q_k = \frac{kp_k}{\langle k \rangle} \,. \tag{2.4}$$

In addition, one can also evaluate the probability that a randomly chosen link has nodes of degree j,k at either end. This is the node joint degree distribution, denoted e_{jk}. Once again this is normalised and satisfies the following relationships:

$$\sum_{jk} e_{jk} = 1, \tag{2.5}$$

$$\sum_{jk} e_{jk} = q_j \tag{2.6}$$

$$e_{jk} = e_{kj} \,, \tag{2.7}$$

where the final result arises from reciprocal nature of the links in an undirected network. As an example, consider again the network in figure 2.1. This has the following probability distributions:

$$\mathbf{p} = \frac{1}{16} \begin{bmatrix} 4 \\ 8 \\ 4 \end{bmatrix} \begin{matrix} 5 \\ 6 \\ 7 \end{matrix} \qquad \mathbf{q} = \frac{1}{96} \begin{bmatrix} 20 \\ 48 \\ 28 \end{bmatrix} \begin{matrix} 5 \\ 6 \\ 7 \end{matrix} \qquad \mathbf{e} = \frac{1}{96} \begin{matrix} \begin{matrix} 5 & \ 6 & \ 7 \end{matrix} \\ \begin{bmatrix} 2 & 9 & 9 \\ 9 & 22 & 17 \\ 9 & 17 & 2 \end{bmatrix} \end{matrix} \begin{matrix} 5 \\ 6 \\ 7 \end{matrix} \tag{2.8}$$

2.1.2 Atomic and Ring Networks

To see how this relates to atomic materials, consider the amorphous graphene configuration in figure 2.2a. In this network the nodes represent carbon atoms and the links sp^2 bonds. The node degree in the atomic network for all nodes is then equal

to three, being equivalent to the atomic coordination number (which throughout this thesis will be denoted by c). This is problematic, because whilst there is clear disorder in the system, it is not well captured by the atomic network. Due to the fact that the local environment around the atoms is identical, when examining say the node degree distribution any information about the glassy structure is lost. This network is to first order indeterminable from a crystalline hexagonal lattice.
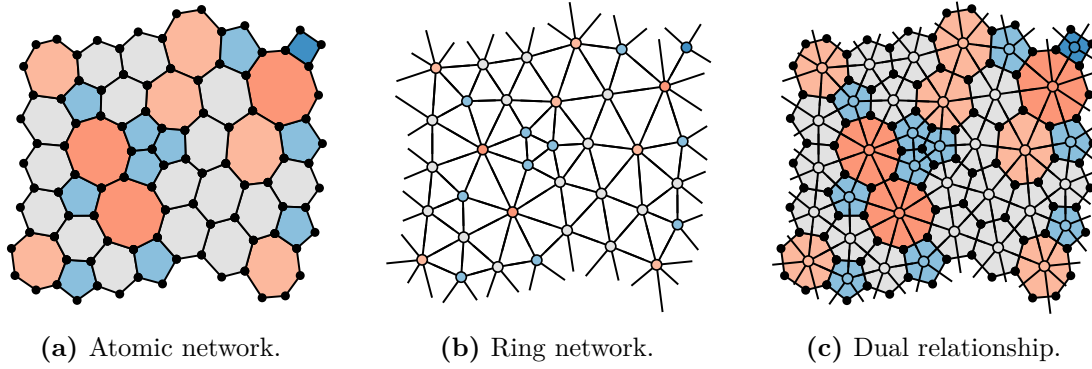

(a) Atomic network.   **(b)** Ring network.   **(c)** Dual relationship.

Figure 2.2: Panel (a) gives an example of a 3- coordinate periodic atomic network with disordered ring structure. Nodes and links represent atoms and bonds respectively where rings are coloured by size. Panel (b) gives the corresponding ring network where nodes and links represent rings and their adjacencies, where nodes are coloured by degree. Panel (c) shows the dual relationship between the atomic and ring networks, where the node degree in the ring network is equal to the ring size in the atomic network.

Observing figure 2.2a one can see there is another level of structure in the network, namely that of the ring structure. A ring is strictly any closed path of sequentially linked nodes in a network, but this thesis will use the term in reference only to the primitive rings *i.e.* those which cannot be subdivided into two smaller rings [50]. A ring of size k (or k-ring) is then defined as a ring with k constituent nodes. It is clear that finding and counting the number of rings of each size, often termed calculating the ring statistics, does then quantify the disorder in the system [27]. The ring statistics can be summarised by the normalised probability distribution, p_k.

However, there is a more efficient way of representing and quantifying the ring structure in the system, and that is by constructing the dual network [51]. The dual is generated by placing a node at the centre of each ring and linking the nodes of adjacent (*i.e.* edge-sharing) rings, as can be seen in figure 2.2b. This will be

referred to as the ring network. The ring network is a reciprocal lattice in which the node degree, k, is equivalent to the ring size in the atomic network. Similarly, it consists solely of triangles, reflecting the 3-coordinate nature of the underlying atomic network. Hence, the disorder is captured directly in the node properties of the ring network. These characteristics make the ring network preferable for manipulating and analysing the systems in this thesis.

2.2 Topological Laws

There are a number of laws which govern the topological properties of two-dimensional network-forming materials. These laws constrain the ring structure, influencing the network properties in a manner that makes physical networks unique in the field of network science. These laws act on a number of "levels": Euler's law controls the overall mean ring size, Lemâitre's law the ring size distribution and the Aboav-Weaire law the ring-ring correlations.

2.2.1 Euler's Law

Euler's law constrains the mean ring size, $\langle k \rangle = \sum_k k p_k$, in an atomic network; equivalent to the mean node degree of the ring network. The atomic networks studied in this work are all two-dimensional, connected (there is a path between any two nodes) and planar (they have no overlapping links) and so are subject to Euler's formula which states:

$$N + V - E = \chi, \tag{2.9}$$

where N, V, E are the number of rings, vertices and edges in the network and χ in an integer termed the Euler characteristic, which is dependent on the global topology of the system. Each vertex represents an atom and the number of edges emanating from each vertex is then the coordination number.

For generality consider an atomic network with atoms of assorted coordination numbers, c. If the proportion of each coordination type is x_c, then the mean coordination number is given by $\langle c \rangle = \sum_c c x_c$. This allows the number of edges

to be written in terms of the number of vertices as $E = \frac{V}{2}\langle c \rangle$. In turn the mean ring size is simply the total number of vertices per ring, allowing for multiple counting, such that $\langle k \rangle = \frac{V}{N}\langle c \rangle$. Substituting these two expressions into equation (2.8) leads to the expression:

$$\langle k \rangle = \frac{2\langle c \rangle \left(1 - \chi/N \right)}{\langle c \rangle - 2}. \tag{2.10}$$

Hence the average node degree in the ring network (equivalent to the mean ring size of the physical network), is simply related to the average degree of the physical network (*i.e.* local coordination environment), the topology of the system and the number of rings.

Although equation (2.9) may appear simple, it is a very powerful constraint. To demonstrate this consider a two-dimensional lattice with two possible coordination environments $c = 3, 4$. The planar case with periodic boundary conditions (mimicking an infinite planar lattice) maps onto the torus with $\chi = 0$, and so:

$$\langle k \rangle = \begin{cases} 6, & x_3 = 1 \\ 4, & x_4 = 1 \\ 5, & x_3 = 2/3, x_4 = 1/3. \end{cases} \tag{2.11}$$

To reiterate in plain terms, this means that if there is a material consisting of atoms all forming exactly three bonds (as for amorphous carbon), the mean ring size *must* be equal to six. Similarly if all atoms form four bonds the mean ring size is four, and if there is a two-thirds to one-third mixture of coordination environments the mean ring size is five. The simplest illustrations of these are the hexagonal, square and cairo regular tilings, shown in figure 2.3, but this law holds equally well for amorphous configurations. For aperiodic systems strictly $\chi = 1$, but as N grows and the proportion of vertices with unsatisfied coordination on the sample perimeter become negligible overall, in reality these relationships hold, and remain as applicable to amorphous graphene as the basalt columns in Fingal's Cave, and the Penrose tiling [34, 52].

This analysis also extends to spherical topology where $\chi = 2$, and so:

$$\langle k \rangle = \begin{cases} \frac{6N-12}{N}, & x_3 = 1 \\ \frac{4N-8}{N}, & x_4 = 1. \end{cases} \tag{2.12}$$

These relationships are the origin of the 12 pentagon rule for 3-coordinate fullerenes (the "football problem"), or equivalently an "8 triangle rule" in the 4-coordinate case, as this is the only way to satisfy these equations if the allowed ring sizes are limited to $k = 5, 6$ and $k = 3, 4$ respectively (as in figures 2.3d,2.3e) [53]. Much of the richness in the behaviour of two-dimensional physical networks stems from this fundamental constraint on the network average degree.
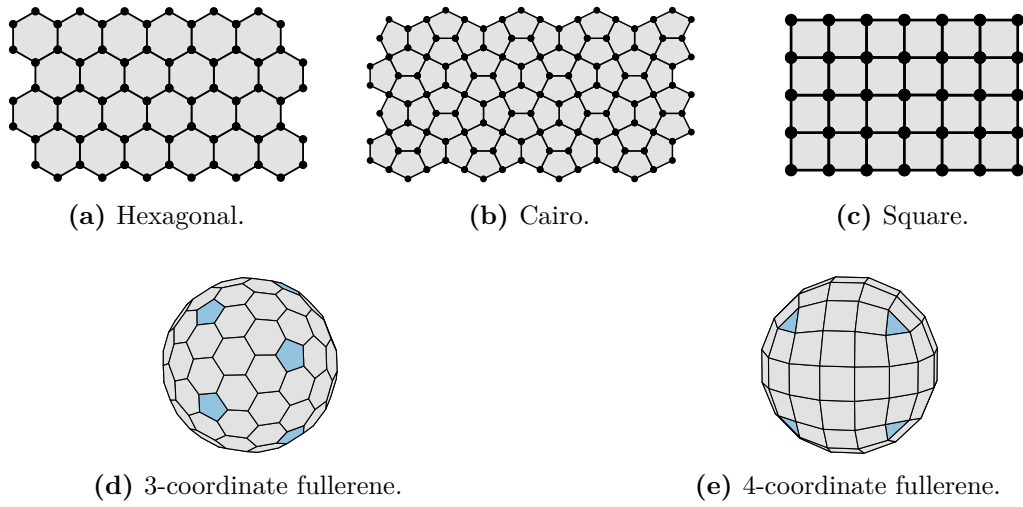

(a) Hexagonal. **(b)** Cairo. **(c)** Square.

(d) 3-coordinate fullerene. **(e)** 4-coordinate fullerene.

Figure 2.3: Panels (a)-(c) give regular planar tilings of 6-, 5- and 4- rings, where the ring size is related to the underlying atomic coordination. Panels (d) and (e) show the 3- and 4- coordinate tilings in spherical topology, where the mean ring size is reduced due to the change in the Euler characteristic.

2.2.2 Lemâitre's Law

Knowing that the mean node degree is fixed by Euler's law, the next level of available information is the form of the underlying degree distribution, p_k. Interestingly, the degree distributions found in physical ring networks seem relatively well defined. For instance, it has been noted in models and realisations of two-dimensional silica glass that the ring statistics looked to follow a lognormal distribution [9, 13]. Lemâitre *et al.* demonstrated that the distribution in 3-coordinate networks systems can be well described by a maximum entropy distribution [54]. Lemâitre's maximum entropy method is summarised here, trivially extended to arbitrary coordination.

The entropy of a probability distribution is defined as $S = -\sum_k p_k \log p_k$. In addition, the degree distribution has the following constraints:

$$\sum_k p_k = 1, \tag{2.13}$$

$$\sum_k k p_k = \langle k \rangle, \tag{2.14}$$

$$\sum_k p_k/k = \text{constant}, \tag{2.15}$$

where the first two constraints correspond to the normalisation condition and the fixed mean ring size, and the final constraint will be discussed below. The entropy can then be maximised using Lagrange's method of undetermined multipliers to yield the result:

$$p_k = \frac{e^{-\lambda_1 k - \lambda_2/k}}{\sum\limits_k e^{-\lambda_1 k - \lambda_2/k}}, \tag{2.16}$$

which can be solved numerically by substitution into equations (2.13),(2.14). By allowing the chosen constant to vary, a family of maximum entropy curves can be generated, as in figure 2.4a. The resulting distributions can be summarised by relating the variance, $\mu_2 = \langle k^2 \rangle - \langle k \rangle^2$, to a single chosen node degree probability, leading to the plot known as Lemâitre's law, given in figure 2.4b. It is usually framed in the context of the proportion of hexagons in a system, p_6, for the precise reason that most networks have $\langle k \rangle = 6$ and p_6 as the largest contribution. Many experimental and theoretical studies have shown good agreement to this law [55–57]. Simple extensions of the classic law are however possible, by modifying the mean degree or the permitted degree range. For instance, k is usually taken in the interval $k \geq 3$ (as the triangle, $k = 3$, is the smallest polygon), but there can be manifestations of physical systems where only certain degrees are accessible [58]. Link to procrystal chapter . The resulting Lemâitre curves for a selection of these modifications are given in figure 2.4c. explain these here or later?   The maximum value of these curves can be simply determined by removing constraint (2.14), equivalent to setting $\lambda_2 = 0$ in equation (2.15).

The only somewhat puzzling aspect of this successful theory is the choice of constraint (2.14). It was originally rationalised on the basis that the areas of rings
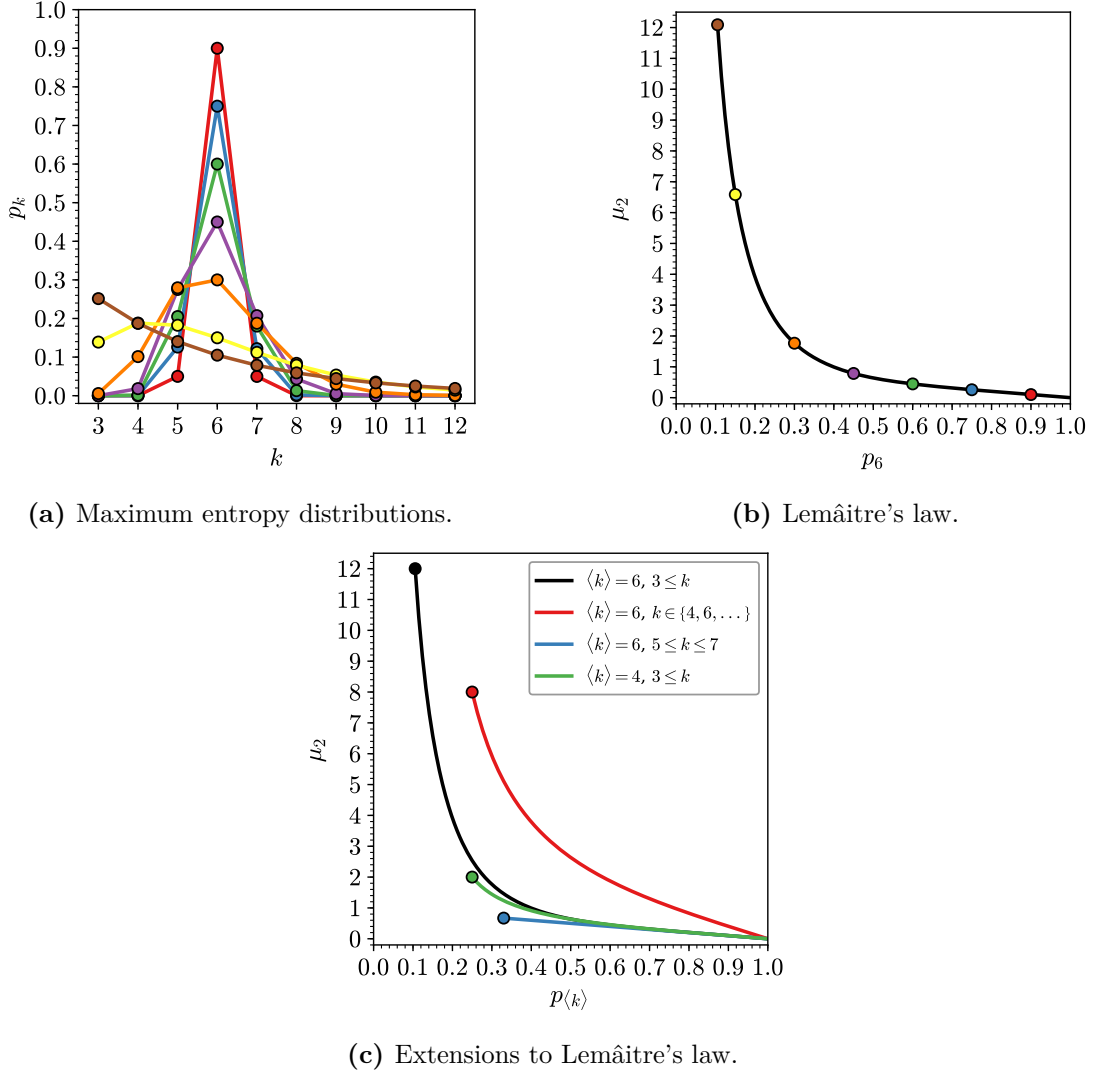
(a) Maximum entropy distributions.

(b) Lemâitre's law.

(c) Extensions to Lemâitre's law.

Figure 2.4: Illustration of Lemâitre's maximum entropy method. Panel (a) gives examples of explicit maximum entropy distributions with different values of p_6. Panel (b) shows how these distributions can be summarised in a plot of p_6 *vs.* μ_2 (Lemâitre's law). Panel (c) provides extensions to the law by modifying the underlying constraints of the mean ring size and allowable k-range.

of a given size, A_k, can be well fit by an expression $A_k = ak + b + c/k$, where a, b and c are constants. As noted at the time, this is by no means true for all systems and in fact is contrary to the widely known Lewis law, which states that A_k is linear in k for many observable networks [59–61]. Despite this, the universality of the Lemâitre law suggests that there must be a physical basis to (2.14), and in the section   Link to later networks   it will be demonstrated that it can be regenerated by considering ring adjacencies.

2.2.3   Aboav-Weaire Law

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