ARTICLES

The von Neumann relation generalized to coarsening of three-dimensional microstructures

Robert D. MacPherson[1] & David J. Srolovitz[2]

Cellular structures or tessellations are ubiquitous in nature. Metals and ceramics commonly consist of space-filling arrays of single-crystal grains separated by a network of grain boundaries, and foams (froths) are networks of gas-filled bubbles separated by liquid walls. Cellular structures also occur in biological tissue, and in magnetic, ferroelectric and complex fluid contexts. In many situations, the cell/grain/bubble walls move under the influence of their surface tension (capillarity), with a velocity proportional to their mean curvature. As a result, the cells evolve and the structure coarsens. Over 50 years ago, von Neumann derived an exact formula for the growth rate of a cell in a two-dimensional cellular structure (using the relation between wall velocity and mean curvature, the fact that three domain walls meet at 120° and basic topology). This forms the basis of modern grain growth theory. Here we present an exact and much-sought extension of this result into three (and higher) dimensions. The present results may lead to the development of predictive models for capillarity-driven microstructure evolution in a wide range of industrial and commercial processing scenarios—such as the heat treatment of metals, or even controlling the 'head' on a pint of beer.

We consider physical systems in which space is divided into cells/grains/bubbles, which we refer to here as 'domains'. We assume that the growth of these domains is driven by capillarity. This applies to grains in metals or ceramics, and to bubbles in foams[1,2]. The rate of change of the volume of a domain is given by a physical constant $-M\gamma$ times the integral of the mean curvature $\mathcal{K}$ over the walls of the domain[3,4]. Here γ is the surface tension of the domain wall and M is a kinetic coefficient that describes its mobility. If the domains are grains in a metal or a ceramic, then a wall between two adjacent domains moves with velocity $-M\gamma\mathcal{K}$ (the Gibbs–Thompson effect)[5]. If the domains are bubbles in a foam, then $-\gamma\mathcal{K}$ is the pressure difference between adjacent bubbles, and the gas diffuses through the wall at a rate $-M\gamma\mathcal{K}$ per unit area, where M is now proportional to the diffusivity[6].

The integral of the mean curvature over all of the walls of a domain is a difficult quantity to use in theories for domain growth. In 1952, von Neumann[1] found a manageable expression for the growth of domains in two-dimensional space:

$$\frac{\mathrm{d}A}{\mathrm{d}t} = -M\gamma\left(2\pi - \sum_{i=1}^{n}\alpha_i\right) = -2\pi M\gamma\left(1-\frac{1}{6}n\right) \qquad (1)$$

Here $\mathrm{d}A/\mathrm{d}t$ is the rate of change of the area of a domain, α_i is the exterior (turning) angle at a triple junction on that domain (where three domain walls meet—see Fig. 1a), and n is the number of such triple junctions around a particular domain. A more general derivation of the same result was provided by Mullins[3]. The derivation of the von Neumann–Mullins formula is this: the domain is surrounded by a closed curve, the total integral of the (mean) curvature $\mathcal{K}$ around which (and indeed around any closed curve) is 2π. However, α_i of this total curvature is localized in an abrupt turn at the ith triple junction, so it does not contribute to the integral of $\mathcal{K}$ along the walls. If the surface tension γ is the same on each wall, the equilibrium angle is $\alpha_i = 2\pi/6$. Like von Neumann and Mullins[1,3], we explicitly assume that the domain walls move sufficiently slowly that this angle remains fixed at its equilibrium value. We treat $\alpha_i = 2\pi/6$ as a boundary condition for three domain walls meeting at a junction (in any dimension ≥ 2).

The von Neumann–Mullins relation is remarkable in that it is both exact and purely topological; the rate of growth of any domain only depends on n, its number of sides (or corners where three domains meet, that is, triple junctions)[1,3]. This relation forms the basis of much of what is known today about isotropic domain growth. Fifty years ago, the grain growth pioneer Smith wrote "It is greatly to be hoped that he [von Neumann], or some other mathematician, will be able to deduce similar relations in three dimensions."[7] Until now, no exact extension of von Neumann's two-dimensional result into three (or higher) dimensions has been found (despite a half century of intense effort in the materials, physics, engineering and mathematics communities[8–13]). The purpose of this Article is to provide an exact extension of this relation into all dimensions, and show that the von Neumann–Mullins relation, and its physically important extension to three dimensions, are simply special cases of the more general mathematical result.

Our three-dimensional von Neumann–Mullins relation is this: the rate of change of the volume V of a domain $\mathbf{D}$ is given by:

$$\frac{\mathrm{d}V}{\mathrm{d}t} = -2\pi M\gamma\left(\mathcal{L}(\mathbf{D}) - \frac{1}{6}\sum_{i=1}^{n} e_i(\mathbf{D})\right) \qquad (2)$$

The quantity $\mathcal{L}(\mathbf{D})$ is a natural measure of the linear size of domain $\mathbf{D}$ and $\sum_{i}^{n} e_i(\mathbf{D})$, where e_i is the length of triple line (edge) i and the summation is over all n triple lines of $\mathbf{D}$ (see Fig. 2a). This result is exact. A proof is provided in the Supplementary Information.

We digress to give the definition and properties of the quantity $\mathcal{L}(\mathbf{D})$. Consider a line, ℓ, through the origin of some coordinate

[1]School of Mathematics, Institute for Advanced Study, Princeton, New Jersey 08540, USA. [2]Department of Physics, Yeshiva University, New York, New York 10033, USA.

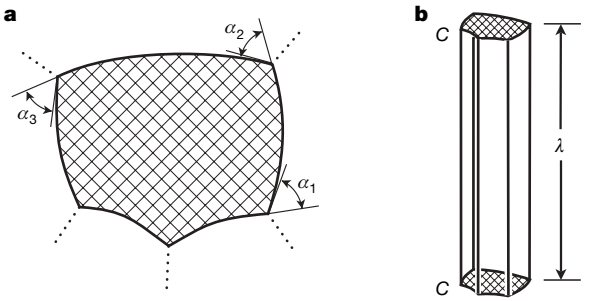

Figure 1 | A two-dimensional grain and its extension into three dimensions. **a**, A schematic illustration of a grain in a two-dimensional network structure. The dotted lines indicate domain boundaries of adjacent domains, and α_i is the turning angle at triple point *i*. **b**, The same grain extended into a three-dimensional prism of length λ and cross-sectional shape *C*.

system in three dimensions; p is a point on ℓ, and $\ell_p^\perp$ is the plane perpendicular to line ℓ at point p. We define the Euler width of a domain $\mathbf{D}$ in the direction ℓ as $E_\ell(\mathbf{D}) = \int_p \chi(\ell_p^\perp \cap \mathbf{D})\mathrm{d}p$ (see Fig. 2b), where the integral is over all points p along line ℓ and $\chi(\ell_p^\perp \cap \mathbf{D})$ is the Euler characteristic of the intersections of the domain $\mathbf{D}$ with plane $\ell_p^\perp$. The Euler characteristic is the number of objects or pieces in the plane $\ell_p^\perp \cap \mathbf{D}$ minus the number of holes in those objects/pieces. (For a convex object with no holes, $\chi = 1$.) The mean width of $\mathbf{D}$, $\mathcal{L}(\mathbf{D})$, is twice the Euler width, averaged over all lines ℓ through the origin. If the object $\mathbf{D}$ is a line or a curve, the mean width is its length; for a convex object, the mean width is twice the average length of the projection onto a line ℓ, so for a sphere it is twice its diameter. The mean width is additive in the sense that if $\mathbf{D}_1$ and $\mathbf{D}_2$ are objects with mean width $\mathcal{L}(\mathbf{D}_1)$ and $\mathcal{L}(\mathbf{D}_2)$, the union of these two objects has mean width $\mathcal{L}(\mathbf{D}_1 \cup \mathbf{D}_2) = \mathcal{L}(\mathbf{D}_1) + \mathcal{L}(\mathbf{D}_2) - \mathcal{L}(\mathbf{D}_1 \cap \mathbf{D}_2)$. Note that the same additivity rule applies to the volume or surface area of a pair of objects. Hadwiger's theorem states that any measure of the linear dimension of a convex body that is additive and continuous is simply proportional to the mean width[14–18]. The mean width of an object can be computed analytically for many shapes, including all flat-faced polyhedra, and computed numerically for arbitrary shapes, as is shown in detail in Supplementary Information.

A few comments on the main result, equation (2), are in order. First, unlike the von Neumann–Mullins result in two dimensions, this result is not purely topological. The rate of change of the domain volume depends on the mean width of the domain and the total length of the triple lines. It does not, however, depend explicitly on grain shape. Note that the summation of the lengths of all triple lines (in equation (2)) can be described as the mean width of the set of triple lines, $\mathcal{L}(\mathrm{edge}(\mathbf{D}))$, where $\mathrm{edge}(\mathbf{D})$ is the set of triple lines.

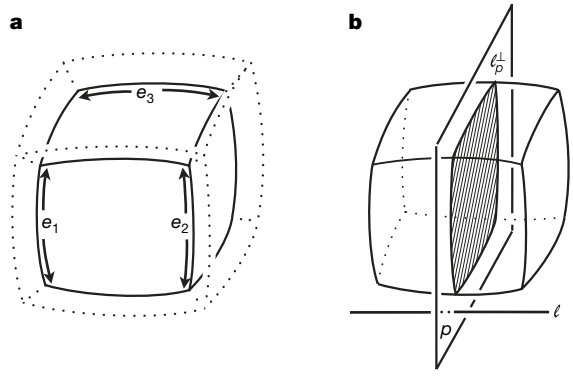

Figure 2 | Notation used in description of the three-dimensional von Neumann–Mullins relation. **a**, A grain with six faces and twelve edges from a three-dimensional network. e_i are the lengths of edge *i*. **b**, The intersection of a domain ($\mathbf{D}$) and plane $\ell_p^\perp$ which has its normal parallel to ℓ at point p.

We can rewrite the right side of equation (2) as $-2\pi M\gamma\mathcal{L}(\mathbf{D})(1 - f/6)$, where $f = \mathcal{L}(\mathrm{edge}(\mathbf{D}))/\mathcal{L}(\mathbf{D})$. If we write the number of faces on a domain (a face is a region of the surface of $\mathbf{D}$ bounded by triple lines) as m, then it can be seen that f scales as $m^{1/2}$. Using this relation, we can rewrite equation (2) approximately as $\mathrm{d}D^2/\mathrm{d}t = C_1 M\gamma(6 - C_2 m^{1/2})$, where we have assumed that all lengths are proportional to the same linear dimension D of domain $\mathbf{D}$, and C_1 and C_2 are constants. This result looks very similar to the classic two-dimensional von Neumann–Mullins result (a similar result was found by Hilgenfeldt *et al.*[10]), that is, the right side of the equation is topological. Unlike the exact extension of the von Neumann relation to three dimensions (equation (2)), this result is approximate and simply shows the correct scaling. As both of the terms on the right side of equation (2) are proportional to the linear dimension of the domain, it can be seen that $\mathrm{d}D^3/\mathrm{d}t$ is simply proportional to D or that $D \propto t^{1/2}$. Such parabolic growth is typical of capillarity-driven domain coarsening[19,20].

We can deduce the two-dimensional von Neumann–Mullins relation from equation (2) by considering a prism of cross-sectional shape C with n sides and length λ in three dimensions (see Fig. 1b). For large λ (holding C constant), the prism appears as a line such that $\mathcal{L}(\mathbf{D}) \approx \lambda$ and the sum of the edge lengths is simply $n\lambda$. Inserting these into equation (2) yields $\mathrm{d}V/\mathrm{d}t \approx \lambda \mathrm{d}A/\mathrm{d}t \approx -2\pi M\gamma(\lambda - \lambda n/6)$. Cancelling λ from each side and taking the limit that $\lambda \to \infty$ exactly yields the two-dimensional von Neumann–Mullins relation.

The von Neumann–Mullins relation can be restated by noting that $\mathrm{d}A/\mathrm{d}t = -(\pi M\gamma/3)(6 - n) = -2\pi M\gamma[\chi(\mathbf{D} - \chi(\mathrm{vert}(\mathbf{D}))/\mathbf{6}]$, where $\chi(\mathrm{vert}(\mathbf{D}))$ is the Euler characteristic of the vertices of the domain $\mathbf{D}$ (that is, the number of triple points). We note that this form of the von Neumann–Mullins relation is very similar to our three-dimensional result written in the form $\mathrm{d}V/\mathrm{d}t = -2\pi M\gamma(\mathcal{L}(\mathbf{D}) - \mathcal{L}(\mathrm{edge}(\mathbf{D}))/6))$. The similarity between the two suggests that there may be a more general expression that works in all dimensions. This is, in fact, true and can be expressed as:

$$\frac{\mathrm{d}V_d}{\mathrm{d}t} = -2\pi M\gamma\left(H_{d-2}(\mathbf{D}_d) - \frac{1}{6}H_{d-2}(\mathbf{D}_{d-2})\right) \qquad (3)$$

where d is the dimension of space, V_d is the volume of domain $\mathbf{D}_d$ in d dimensions, and $\mathbf{D}_{d-2}$ is the $(d-2)$-dimensional feature of the domain (for example, vertices in two dimensions, edges in three dimensions, …). In this expression, H_{d-2} is known as the Hadwiger $(d-2)$-measure in geometric probability[14,15]. The definition of H is akin to that of $\mathcal{L}$ above. Consider a $(d-2)$-dimensional plane, ℓ, through the origin in d dimensions; p is a point on ℓ, and $\ell_p^\perp$ defines the two-dimensional plane perpendicular to ℓ at p. $H_{d-2}(\mathbf{D}_d)$ is equal to the average of $E(\mathbf{D}_d) = \int_p \chi(\ell_p^\perp \cap \mathbf{D}_d)\mathrm{d}p$ over all planes ℓ through the origin. In two dimensions, $H_0(D_2) = \chi(D_2)$ and $H_0(D_0) = \chi(\mathrm{vert}(D))$. In three dimensions, $H_1(\mathbf{D}_3) = \mathcal{L}(\mathbf{D}_3)$ and $H_1(\mathbf{D}_1) = \mathcal{L}(\mathrm{edge}(\mathbf{D}_3))$. These agree with the two-dimensional von Neumann relation and our exact three-dimensional result, respectively.

As most modern models for the evolution of polycrystalline microstructures start with a postulated extension of the two-dimensional von Neumann–Mullins result into three dimensions, the new, exact theory for the evolution of each grain provides a firm foundation for the development of rigorous statistical models for microstructure evolution that respect the underlying geometric and topological constraints of a space-filling network[21,22].

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1. von Neumann, J. in *Metal Interfaces* (ed. Herring, C.) 108–110 (American Society for Metals, Cleveland, 1952).
2. Mullins, W. W. in *Metal Surfaces: Structure, Energetics, and Kinetics* (eds Robertson, W. D. & Gjostein, N. A.) 17–66 (American Society for Metals, Metals Park, Ohio, 1963).
3. Mullins, W. W. Two-dimensional motion of idealized grain boundaries. *J. Appl. Phys.* **27**, 900–904 (1956).

4. Glazier, J. A. Grain growth in three dimensions depends on grain topology. *Phys. Rev. Lett.* **70,** 2170–2173 (1993).
5. Burke, J. E. & Turnbull, D. Recrystallization and grain growth. *Prog. Metal Phys.* **3,** 220–292 (1952).
6. Durian, D. J., Weitz, D. A. & Pine, D. J. Scaling behavior in shaving cream. *Phys. Rev. A* **44,** R7902–R7905 (1991).
7. Smith, C. S. in *Metal Interfaces* (ed. Herring, C.) 65–113 (American Society for Metals, Cleveland, 1952).
8. Rhines, F. N. & Craig, K. R. Mechanism of steady-state grain growth in aluminum. *Metall. Trans.* **5,** 413–425 (1974).
9. Rivier, N. On the structure of random tissues or froths, and their evolution. *Phil. Mag. B* **47,** L45–L49 (1983).
10. Hilgenfeldt, S., Kraynik, A. M., Koehler, S. A. & Stone, H. A. An accurate von Neumann's law for three-dimensional foams. *Phys. Rev. Lett.* **86,** 2685–2688 (2001).
11. Hilgenfeldt, S., Kraynik, A. M., Reinelt, D. A. & Sullivan, J. M. The structure of foam cells: Isotropic plateau polyhedra. *Europhys. Lett.* **67,** 484–490 (2004).
12. Glicksman, M. E. Capillarity-mediated grain growth in 3-D. *Mater. Sci. Forum* **467–470,** 1025–1031 (2004).
13. Glicksman, M. E. Analysis of 3-D network structures. *Phil. Mag.* **85,** 3–31 (2005).
14. Hadwiger, H. *Vorlesungen über Inhalt, Oberfläche und Isoperimetrie* (Springer, Berlin, 1957).
15. Klain, D. A. & Rota, G.-C. *Introduction to Geometric Probability* (Cambridge Univ. Press, Cambridge, UK, 1997).
16. Federer, H. Curvature measures. *Trans. Am. Math. Soc.* **93,** 418–491 (1959).
17. Fu, J. H. G. Curvature measures of subanalytic sets. *Am. J. Math.* **116,** 819–880 (1994).
18. Bröcker, L. & Kuppe, M. Integral geometry of tame sets. *Geom. Dedicata* **82,** 285–323 (2000).
19. Atkinson, H. V. Theories of normal growth in pure, single phase systems. *Acta Metall.* **36,** 469–491 (1988).
20. Mullins, W. W. Estimation of the geometrical rate constant in idealized three dimensional grain growth. *Acta Metall.* **37,** 2979–2984 (1989).
21. Hillert, M. On the theory of normal and abnormal grain growth. *Acta Metall.* **13,** 227–238 (1965).
22. Abbruzzese, G., Heckelmann, I. & Lucke, K. Statistical theory of two-dimensional grain growth. *Acta Metall. Mater.* **40,** 519–532 (1992).

Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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