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Wulff construction and anisotropic surface properties of two-dimensional Eden clusters

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Abstract. A relation between the shape of Eden clusters and the number of perimeter sites per unit area of the surface is derived which is analogous to the Wulff construction of equilibrium shapes in thermodynamic systems. New data are presented for the surface width and the surface skewness of Eden clusters grown on a square lattice. The width depends on the average orientation of the surface with respect to the underlying lattice. Its corrections to scaling are discussed. The skewness has unexpected changes of sign.

1. Introduction

Several models simulating on a lattice the growth of a cluster by random aggregation of particles produce anisotropic structures reflecting the underlying lattice symmetry. This has recetly been observed, e.g. for diffusion-limited aggregation (DLA) [1] and for the Eden model [2, 3] on square and simple cubic lattices, which count as prototypes among the growth models (for a review see [4]). In both cases the diameter measured in Euclidean metric on the square lattice and averaged over many independent clusters of equal size is larger along the lattice axes than along the lattice diagonals. However, whereas for DLA they differ by at least 30% the anisotropy is much weaker for Eden clusters on a square lattice, namely about 2% only. Thus the sensitivity with respect to the lattice structure depends on the growth rule.

The growth rule investigated in this paper is Eden model A in the classification of [5]. At every time step a new particle is put on a randomly chosen perimeter site (empty nearest neighbour of an already occupied site in the cluster). The shape anisotropy was observed in [1] and [2] for clusters grown out of a point seed. However, in many respects Eden clusters grown on a flat substrate [5] with well defined orientation with respect to the lattice allow a more systematic investigation of the anisotropic growth properties. The use of a substrate instead of a point seed is natural in the Eden model as the clusters are compact, i.e. there remain no holes deep in the interior of the cluster [6].

The purpose of this paper is twofold. Firstly in § 2 a formula will be derived which connects the shape of an Eden cluster grown out of a point seed with a surface property measured for clusters grown on a flat substrate, namely the orientational dependence of the number N_p of perimeter sites per Euclidean unit length L of the substrate [3]. This quantity can be shown to have the meaning of a growth velocity. The formula is similar to the Wulff construction of equilibrium shapes of crystals [7, 8] from the orientational dependence of the surface tension.

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Secondly, new data are presented in § 3 concerning the perimeter site distribution. They were obtained in a computer simulation of Eden clusters grown on flat substrates with four different orientations WRT the square lattice, namely parallel to a lattice axis (Miller indices (0,1)), parallel to a lattice diagonal (Miller indices (1,1)) and at angles $\tan^{-1} 1/n$ with a lattice axis (Miller indices $(1, n)$) for $n = 2$ and 4. Two quantities have been investigated; the width of the surface region

$$w = \langle (r - \langle r \rangle)^2 \rangle^{1/2} \tag{1.1}$$

and the surface skewness

$$s = \langle (r - \langle r \rangle)^3 \rangle / w^3 \tag{1.2}$$

where r is the Euclidean distance of a perimeter site from the substrate. The angular brackets indicate averaging over all perimeter sites. Both quantities w and s were averaged over a large number of clusters of equal size. For a symmetric probability distribution of the distances r the skewness would be zero. To my knowledge this quantity has not been investigated systematically for Eden cluster surfaces before.

A substrate with Miller indices $(1,n)$ has plateaux of width n parallel to the x axis of the lattice. These are separated by steps of height one in the y direction. The cluster is then grown in an infinite strip in y direction, the width X of which (measured parallel to the x axis) is a multiple of the plateau width n. Along the edges of this strip periodic boundary conditions are imposed so that sites on opposite edges are identified which have equal distance from the substrate. This can be handled most conveniently if the Miller indices are not exactly $(1,n) = (X/n, X)$ but $(X/n - 1, X)$. The error one makes is very small for $X \gg n$. Therefore I shall not distinguish in the following which data have been obtained with the exact or with the approximate Miller indices. For Miller indices $(1,n)$ the substrate length L is related to X by

$$L = X(1 + n^2)^{1/2}/n. \tag{1.3}$$

Finally in § 4 the results will be summarised.

2. Wulff construction for Eden clusters

In this paper the shape of Eden clusters is understood in an average sense. Consider a large number of clusters of equal size grown from a point seed at the origin of the coordinate system. For every lattice cell we average the occupation number (0 if the cell is empty, 1 if it belongs to the cluster) over all clusters. This gives a density function $d(\boldsymbol{r},t)$, where the time of growth t corresponds to the cluster size. For the moment let us define the position of the surface, $\boldsymbol{r}(p, t)$, as a contour of constant density close to the maximum gradient, with a parametrisation p.

Suppose that Eden clusters develop a stationary shape evolving in time just through a scaling of the linear dimension. Then one can choose the parametrisation such that

$$\boldsymbol{r}(p, t) = \bar{r}(t)\boldsymbol{r}_0(p) \tag{2.1}$$

with a normalised shape $\boldsymbol{r}_0(p)$. For the normal growth velocity this implies

$$v(p, t) = (\mathrm{d}\boldsymbol{r}/\mathrm{d}t) \cdot \boldsymbol{n} = \bar{v}(t) v_0(p) \tag{2.2}$$

where $\boldsymbol{n} = \boldsymbol{n}(p)$ is the surface normal and $\bar{v}$ and v_0 are given by

$$\begin{aligned} \bar{v}(t) &= \mathrm{d}\bar{r}(t)/\mathrm{d}t \\ v_0(p) &= \boldsymbol{r}_0(p) \cdot \boldsymbol{n}(p). \end{aligned} \tag{2.3}$$

Neglecting curvature corrections we assume that the growth velocity is determined by the orientation of the surface, i.e. v_0 depends on p only through $\boldsymbol{n}$. This orientational dependence of the growth velocity causes the shape of the Eden clusters to be anisotropic. How the growth velocity determines the shape was first described by Wulff [7] for the case of faceted crystals. His construction can be generalised to the present case.

In a first step I show that the shape of Eden clusters has to be convex under the above conditions. The normal growth velocity is always positive because occupied sites remain occupied for ever. It follows then from stationarity (2.3) that the region enclosed by $\boldsymbol{r}_0(p)$ has to be starlike. Every point on the curve $\boldsymbol{r}_0(p)$ can be connected with the origin by a straight line lying entirely in the enclosed region. Hence we can choose the polar angle φ as parameter p. Denoting the normal direction by ϑ (figure 1) (2.3) becomes

$$v_0(\vartheta(\varphi)) = r_0(\varphi)\cos(\varphi - \vartheta(\varphi)). \tag{2.4}$$

An elementary geometrical consideration shows that the normal direction is given by

$$\tan(\varphi - \vartheta(\varphi)) = r_0'(\varphi)/r_0(\varphi). \tag{2.5}$$

Inserting the φ derivative of (2.4) in (2.5) one obtains

$$0 = [\tan(\varphi - \vartheta) - v_0'(\vartheta)/v_0(\vartheta)]\vartheta'(\varphi). \tag{2.6}$$

Therefore the surface is either planar ($\vartheta'(\varphi) = 0$) or

$$\tan(\varphi - \vartheta) = v_0'(\vartheta)/v_0(\vartheta). \tag{2.7}$$

Because (2.3) is positive ($\varphi - \vartheta$) lies between $-\pi/2$ and $\pi/2$, so that (2.7) can uniquely be solved for φ. This means that the average shape of Eden clusters is convex, as there exists only one polar angle for which the normal points in a given direction.

Using this result one can now write (2.4) in the form

$$v_0(\vartheta) = \max_{\varphi}[r_0(\varphi)\cos(\varphi - \vartheta)]. \tag{2.8}$$

The maximum condition specifies the polar angle φ under which the vector normal to the surface has a diirection given by the angle ϑ. It follows from convexity (see figure 1). As the tangent at φ has to lie entirely outside the cluster one concludes that

$$r_0(\tilde{\varphi})\cos(\tilde{\varphi} - \vartheta) \leq r_0(\varphi(\vartheta))\cos(\varphi(\vartheta) - \vartheta) \quad \forall\tilde{\varphi}.$$

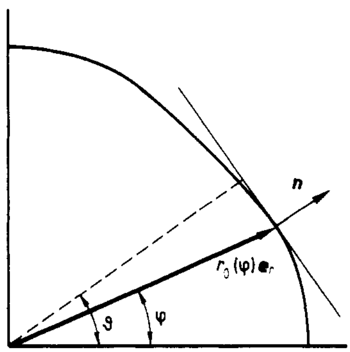

Figure 1. Illustration of the geometrical considerations described in the text to derive the Wulff construction for Eden clusters.

If the orientational dependence $v_0(\vartheta)$ of the growth velocity is known one can now calculate the shape $r_0(\varphi)$ by inverting (2.8):

$$r_0(\varphi) = \min_{\vartheta}\left(\frac{v_0(\vartheta)}{\cos(\varphi-\vartheta)}\right). \tag{2.9}$$

This is completely analogous to the Wulff construction of equilibrium shapes of, for example, Ising domains [8] or crystals [7] where the orientational dependence of the surface tension replaces that of the growth velocity.

One can think of other definitions of the average position of the surface of Eden clusters. In order to make sense they may differ from (2.1) only by order of magnitude of the surface width. Since the width scales with the linear dimension $\bar{r}(t)$ as $\bar{r}^{\beta}$ with $\beta \approx \frac{1}{3}$ [9] the surface region becomes arbitrarily narrow on the scale $\bar{r}(t)$ in the limit $t \to \infty$. Thus all these definitions lead to the same normalised shape $r_0(\varphi)$.

For practical purposes one might prefer a definition such as the following. For every cluster, determine the mean distance from the seed of all perimeter sites within a small solid angle. Average this distance over many clusters of equal size to get $r(\varphi, t)$. However, from a conceptual point of view it is simpler to define a Gibbs surface [10] in the following way: for a small solid angle around a certain direction the true density profile through the surface is replaced by a step function such that the average number of occupied cells within the solid angle remains the same.

It can easily be verified that a surface element of the Gibbs surface propagates with a normal velocity which is equal to the average number of perimeter sites per unit length. At every time step all perimeter sites have equal probability to become occupied. Therefore the deposition rate in a certain direction is equal to the average number of perimeter sites δN_p in a small solid angle $\delta\varphi$ around this direction. Thus the mass deposited in $\delta\varphi$ within a given time interval dt is equal to $\delta N_p\, dt$, where the timescale has been chosen such that in unit time a cluster grows by addition of a number of sites equal to the average total number of perimeter sites. The distance of the surface from the seed thereby increases by $dr = \delta N_p\, dt/(r \cdot \delta\varphi)$. Hence the normal growth velocity (2.2) is asymptotically equal to the number of perimeter sites per unit length of the Gibbs surface

$$v = (dr/dt)\boldsymbol{e}_r \cdot \boldsymbol{n} = \delta N_p/\delta L \tag{2.10}$$

where $\delta L = r\delta\varphi/\boldsymbol{e}_r \cdot \boldsymbol{n}$ is the length of the surface within the solid angle $\delta\varphi$, which must be small enough that one can neglect the curvature of the surface. $\boldsymbol{e}_r$ is the radial unit vector.

The normal growth velocity can be measured for Eden clusters grown on flat substrates. In this case the Gibbs surface is parallel to the substrate and both have the same length. The normal growth velocity differs from the number of perimeter sites per unit length of the substrate only by curvature corrections which can be neglected for large clusters. It has been shown [3] that N_p/L depends on the orientation of the substrate with respect to the underlying square lattice.

Now we can compare the shape anisotropy observed in [2] and the orientational dependence of the normal growth velocity reported in [3]. Notice that figure 2 of [2] was obtained by subtracting a constant from every distance of a perimeter site from the seed in order to exaggerate the small anisotropy. Actually the difference between the axial and diagonal diameters was about 2% which is in good agreement with the difference between the growth velocities given in [3]. For symmetry reasons $\boldsymbol{n}$ has to be parallel to $\boldsymbol{e}_r$ in these directions (i.e. $\varphi = \vartheta$) so that, according to (2.9), the ratio between the axial and diagonal diameters is equal to the ratio between N_p/L for substrates parallel to an axis and to a diagonal. These growth velocities are 2.180 and 2.137, respectively, with errors ±0.007.

3. Surface width and skewness

In order to get a clearer picture about the orientational dependence of perimeter properties in Eden clusters, width and skewness of the perimeter site distribution have been measured in a computer simulation on a CDC Cyber 176. Using single bit handling, and storing only the surface region of a cluster, substrate lengths up to 20 010 were studied. The cluster size is only limited by the computation time but not by memory. The largest clusters contained more than 100 million sites. Intermediate stages of every cluster were evaluated to get data about the time evolution. All results were averaged over a number of independent clusters which varies between 1000 for $L=100$, 100 for $L=1000$ and 50 for $L=20\,000$. The program took about 13 μs per site for the orientations (0,1) and (1,1) and about 15 μs per site for the other orientations.

The width is assumed to scale like [5, 9]

$$w = L^{1/2} f(h/L^z) \tag{3.1}$$

where $h = N/L$ is the distance of the Gibbs surface from the substrate (called the deposit height). Values for z reported in the literature are close to $\frac{3}{2}$ [11, 5, 9]. Plotting $w/L^{1/2}$ against $h/L^{3/2}$ I found a rough collapse of the data for L between 90 and 20 010. The width increases monotonically until it reaches a stationary value around $h = 0.4\,L^{3/2}$. A more detailed analysis of the time evolution of w has been given in [3], where also the strong corrections to scaling are discussed which make it extremely difficult to determine z for Eden model A. The stationary values of $w/L^{1/2}$ are plotted in figure 2 for different substrate lengths and substrate orientations.

According to (3.1) the asymptotic values of $w/L^{1/2}$ should not depend on L. However, figure 2 shows that there are strong corrections to scaling below $L=500$. In this connection it is worth mentioning that for small L the surface width reaches its stationary value before the perimeter to length ratio N_p/L does, whereas for large L

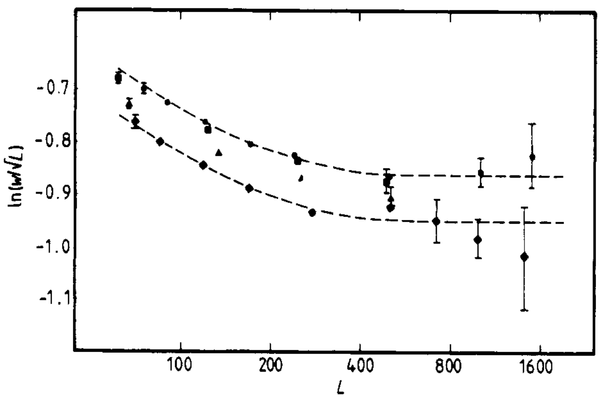

Figure 2. Stationary values of the surface width w scaled by the square root of the substrate length L for Miller indices (0, 1), ●; (1, 4), ■; (1, 2), ▲; (1, 1), ◆.

it is the other way around. The time evolution of N_p/L is given by

$$N_p/L = g(h) \tag{3.2}$$

and the deposit height $h \sim 2000$ where N_p/L reaches its stationary value does not depend on L in contrast to (3.1) [3]. The crossover between the two L regimes happens around $L = 300$ which follows from $2000 = 0.4\, L^{3/2}$. Of course, to attempt an explanation of the corrections to scaling for w by some interaction between the relaxation processes for N_p/L and w is highly speculative but might inspire further work. In this respect a comparison with Eden models B and C where some corrections to scaling are less pronounced [5] could be very instructive.

Figure 2 clearly shows the anisotropy of the surface width. It is largest for surfaces with an average orientation parallel to a lattice axis and decreases if the surface is tilted towards the lattice diagonal. The curves for Miller indices (0,1) and (1,1) are parallel within the error bars in the logarithmic plot of figure 2. This is indicated by the broken lines which, in the horizontal part, are the weighted average of all the data for $L > 490$. It means that the ratio of the stationary values of the surface width for Miller indices (0,1) and (1,1) at the same substrate length L is practically independent of L. This ratio as well as those for the other orientations have been determined for small L where the data are most accurate. Within the error bars they are the same for all substrate lengths considered. Using this observation the values of $w/L^{1/2}$ for large L where the corrections to scaling can be neglected are 0.423, 0.416, 0.405 and 0.388 with errors ± 0.006 for Miller indices (0, 1), (1, 4), (1, 2) and (1, 1), respectively. The anisotropy is stronger than for N_p/L: the difference between $w/L^{1/2}$ for (0, 1) and (1, 1) is about 9% whereas for N_p/L it is about 2% [3].

The surface skewness $\langle (r - \langle r \rangle)^3 \rangle / w^3$ plotted in figure 3 for Miller indices (0, 1) shows a very surprising behaviour as a function of both the substrate length L and the deposit height h. One can understand that it is positive for small h as the perimeter site distribution is truncated by the substrate. That it becomes negative as h increases may be due to the fact that the Eden growth rule allows isolated holes in the cluster but no isolated 'islands' outside, so that one expects that the perimeter site distribution has a longer tail inside the cluster than outside. However, surprisingly for sufficiently

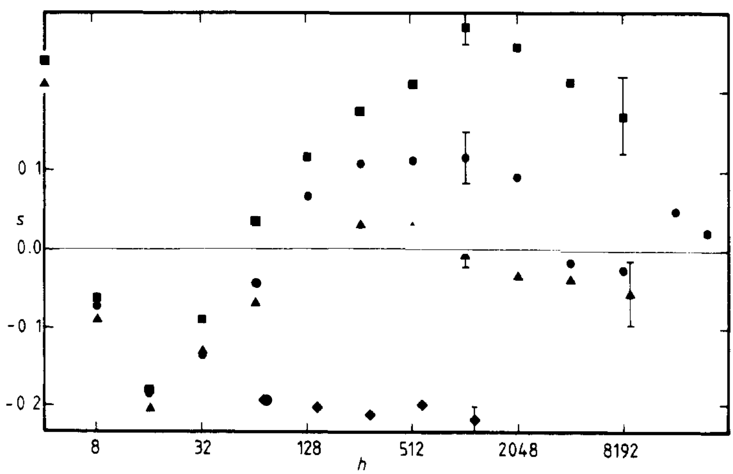

Figure 3. Surface skewness s as function of the deposit height h for Miller indices (0, 1) and substrate lengths $L = 8010$ (■), 1005 (●), 495 (▲) and 120 (◆).

large L the skewness changes sign again, goes through a maximum and maybe oscillates for larger h. The L dependence is very strong. The larger L, the higher is the maximum of s after the second change of sign and the more it is shifted to larger h values. I refrain from speculating on the asymptotic and scaling behaviour of the surface skewness. In particular one has to extend the simulation to larger h values than I did if one wants to investigate its stationary values.

In figure 4 the surface skewness is plotted for Miller indices (1, 1). It shows the same features as a function of L and h as for Miller indices (0, 1). No appreciable anisotropy can be observed for the skewness if one compares the data for Miller indices (0, 1) and $L=495$, $L=1005$ with those for Miller indices (1, 1) and $L=508$, $L=989$. The skewness seems to be slightly smaller for Miller indices (1, 1) than for (0, 1) but more data are needed to make a quantitative analysis. Notice however that s is normalised by the surface width which is different in figures 3 and 4 due to the anisotropy.

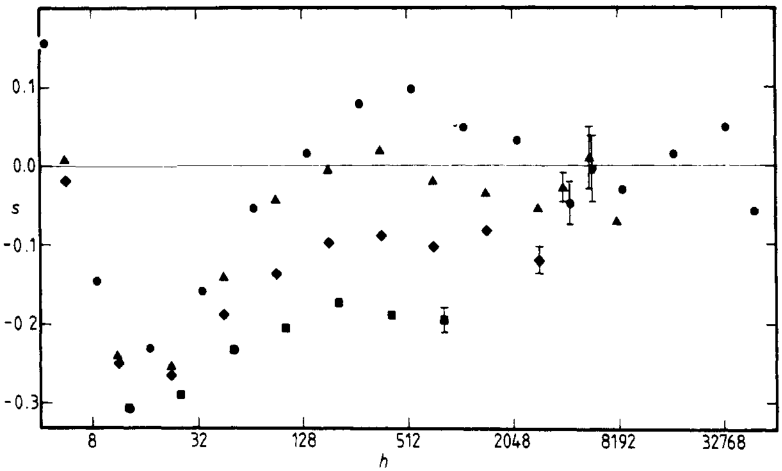

Figure 4. Same as figure 3, but for Miller indices (1, 1) and substrate lengths $L=989$ (●), 508 (▲), 276 (◆) and 170 (■).

4. Conclusion

The main result of this paper is that it is possible to construct the stationary shape of two-dimensional Eden clusters from a Wulff plot of the average number of perimeter sites per unit length of a macroscopically flat surface in exactly the same way as one constructs equilibrium shapes of crystals from a Wulff plot of the surface tension [8]. This result can immediately be generalised to higher dimensions where (2.9) is replaced by

$$r_0(\boldsymbol{e}_r) = \min_{\boldsymbol{n}} \left(\frac{v_0(\boldsymbol{n})}{\boldsymbol{e}_r \cdot \boldsymbol{n}} \right) \tag{4.1}$$

where $v_0(\boldsymbol{n})$ can be identified with the number of perimeter sites per unit area of the Gibbs surface which can be introduced along similar lines as in § 2. It could be shown analytically, by estimating the number of 'histories' leading to a given cluster, that v_0 depends on $\boldsymbol{n}$ at least in space dimensions $d \geqslant 54$ [12]. Numerical evidence for the anisotropy of the growth velocity for $d=2$ was given in [3]. This suggests that Eden clusters may reflect the symmetry of the underlying lattice in all dimensions.

Not only the growth velocity is anisotropic for two-dimensional Eden clusters but also the surface width. Its stationary values in axial and in diagonal direction differ by about 9%. The difference in the growth velocities is only 2%. After completion of this work I received a preprint by Meakin *et al* [13] who report similar results for version C of the Eden model [5]. In their case the growth velocity and the surface width are smaller in the diagonal direction than along the axes by about 2.5% and 11%, respectively. (Notice that for a comparison the values $\sqrt{2}\sigma/\sigma'$ in table 1, [13], have to be multiplied by $2^{1/4}$ in order to get the ratios of width/(substrate length)$^{1/2}$ which I have considered in this paper.)

The surface skewness has a much more complicated behaviour than N_p/L and w. As a function of deposit height its behaviour is clearly not monotonic. Instead it changes sign several times, depending on the substrate length. More data are needed to make a quantitative analysis of its behaviour for large L and h.

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