Dynamics of two-dimensional soap froths

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(Received 12 January 1987)

We have studied experimentally the dynamics of soap froths in two dimensions. We observe two temporal regimes: a transient in which the average bubble area increases in time faster than a power law, and long-term behavior in which the average bubble area increases (over the decade observed) as a power law with exponent $\alpha = 0.59^{+0.11}_{-0.09}$. The crossover between these two regimes depends on the initial preparation. In initially disordered systems the rate of area growth increases smoothly and monotonically, whereas in initially ordered systems the rate first overshoots its long-term value and then decreases. The system does not, however, equilibrate during the lifetime of the bubble lattice. We have also verified that Von Neumann's law for the growth rates of bubbles holds statistically. Lewis's hypothesis of a linear relation between a bubble's number of sides and its area fails for few-sided bubbles. Finally we present a simple phenomenological model for the growth of the average area. This model allows us to define a parameter $\partial(t)$ quantifying the disorder as a function of time.

I. INTRODUCTION

Cellular patterns are common in nature, e.g., magnetic domains in magnetic systems, crystalline domains in ceramics and alloys, biological tissues, etc. The dynamics of such patterns are generally poorly understood.[1]

We have studied two-dimensional soap froths as one of the most experimentally accessible of such cellular patterns. The basic mechanism of evolution is the diffusion of gas across the soap membranes, due to pressure differences between adjacent bubbles.

Smith studied the two-dimensional soap froth as a model for crystal grain growth in metals.[2] He concluded that the average[3] area $\langle a \rangle$ of a bubble increased linearly with time. However, his limited number of observations did not allow definite conclusions. Weaire, working with data collected by Aboav from additional photographs taken by Smith, concluded that $\langle a \rangle$ grows as the square of time.[4-6] Computer simulations also conducted by Weaire have yielded a variety of long-term power laws.[6]

We have studied the rate of area growth in detail and find two distinct temporal regimes: a transient in which $\langle a \rangle$ increases approximately exponentially in time and a long-time tail in which, over the one decade observed (up to 400 h), $\langle a \rangle$ increases as t^α, where $\alpha = 0.59^{+0.11}_{-0.09}$. The nature of the transient depends on initial conditions. For initially well-ordered states, i.e., states in which almost all bubbles have six sides, $d\log\langle a \rangle / d\log(t)$ increases to a value greater than α and then decreases smoothly to α. For initially disordered states $d\log\langle a \rangle / d\log(t)$ increases smoothly and monotonically to α.

We have also checked experimentally two other theoretical predictions, that of Von Neumann and that of Lewis. Von Neumann proposed that the growth rate of a bubble should depend solely on its number of sides, i.e., that $da_i/dt = \kappa(6-n_i)$, where i indexes the ith bubble and n_i its number of sides and κ is a constant.[7] We find that this relation is not true for individual bubbles, but does hold when the rates for all bubbles with a given number of sides are averaged at a fixed time.[8]

Lewis proposed that the area of a polygonal cell should be a linear function of its number of sides.[9] While this relation was originally proposed for biological systems, it has been assumed to hold for two-dimensional soap froths.[1] We find that it fails for few-sided bubbles (i.e., those with three and four sides), though it seems approximately true for many-sided bubbles.

Finally, we propose a model which reproduces the observed pattern of bubble growth and allows us to define the *system disorder*, a parameter $\partial(t)$ ranging from 0 (fully ordered) to 1 (fully disordered) which quantifies the degree of disorder in the system.

II. EXPERIMENTAL PROCEDURE

We performed the experiments in two rectangular Plexiglass cells, one with internal dimensions $6\frac{1}{8} \times 9\frac{3}{16} \times \frac{1}{8}$ in.^{3} (the $\frac{1}{8}$-in. cell) and the other, $7\frac{7}{16} \times 10\frac{1}{8} \times \frac{1}{16}$ in.^{3} (the $\frac{1}{16}$-in. cell). We filled the cell entirely with a soap solution consisting of water (85% by volume), Dawn brand liquid detergent (10%), and glycerol (5%). Our results seem independent of the exact composition of the solution. We next tipped the cell on edge and injected gas bubbles at the bottom of the fluid. Excess fluid drained through a valve at the bottom of the cell. The variation in the size of the injected bubbles determined the initial degree of disorder in the froth. In different runs we used bubbles either of helium or air, helium froths evolving roughly five times faster than air froths, but being otherwise similar. When necessary we "annealed" the filled cell by injecting excess fluid and gently tipping the cell to remove obvious irregularities. We then drained the excess fluid (we did not measure the volume of fluid remaining in the cell), injected a small amount of ink, and sealed the cell with corks and vacuum grease. The ink made the Plateau borders, the thickened region of fluid between the membranes and the walls, easily visible.[1] Finally we placed the

prepared cell level on a photocopier and copied as the rate of evolution required (at intervals of 15 min at early times and 12 h at long times). The photocopies show the Plateau borders and not the soap membranes themselves.

III. RESULTS

In Fig. 1 we present plots of the number of bubbles as a function of time, defined as the total cell area divided by the average area of the bubbles not in contact with the lateral walls of the cell (bulk bubbles). The number of bulk bubbles was counted by hand and the bulk area measured using a digitizing tablet. In Fig. 2(a) we present details (corresponding to 15% of total area) of photocopies of an initially ordered run [the initial disorder $\partial_0 \equiv \partial(0) = 0.17$] taken at the times indicated in Fig. 1(d).

We may qualitatively distinguish these figures as follows.

(A) The bubble lattice is essentially ordered, being composed of hexagonal crystal grains with defects consisting of five- and seven-sided bubbles at the grain boundaries. All bubbles are essentially the same size. Most are six sided. A few are five or seven sided. The rate of evolution is slow.

(B) The grain boundaries become visibly marked as five-sided bubbles shrink and seven-sided bubbles grow. However, six-sided bubbles do not evolve. The number of bubbles with $n \neq 6$ increases, as does the rate of evolution.

(C) The grain boundaries grow into patches of disorder which eat away at the ordered regions. The ordered and disordered regions occupy essentially equal areas. The normalized width of the area distribution ($\langle \delta a \rangle / \langle a \rangle$) is maximal. Many-sided bubbles are common as there is a large probability for a large bubble to be surrounded by much smaller bubbles.

(D) The ordered regions have almost entirely disappeared. The width of the normalized area distribution and the rate of evolution begin to decrease. The number of many-sided bubbles decreases. The fraction of five-sided bubbles, $\rho(5)$, increases monotonically, while the fraction of six-sided bubbles, $\rho(6)$, decreases.

(E) and (F) Long-term states. The evolution rate is essentially constant. There are almost no three-sided bubbles and many-sided bubbles are rare. However, the fraction of bubbles with more than seven sides, $\rho(n)$, $n > 7$, increases slowly. There is no sign of a theoretically predicted collapse in the width of $\rho(n)$.[11]

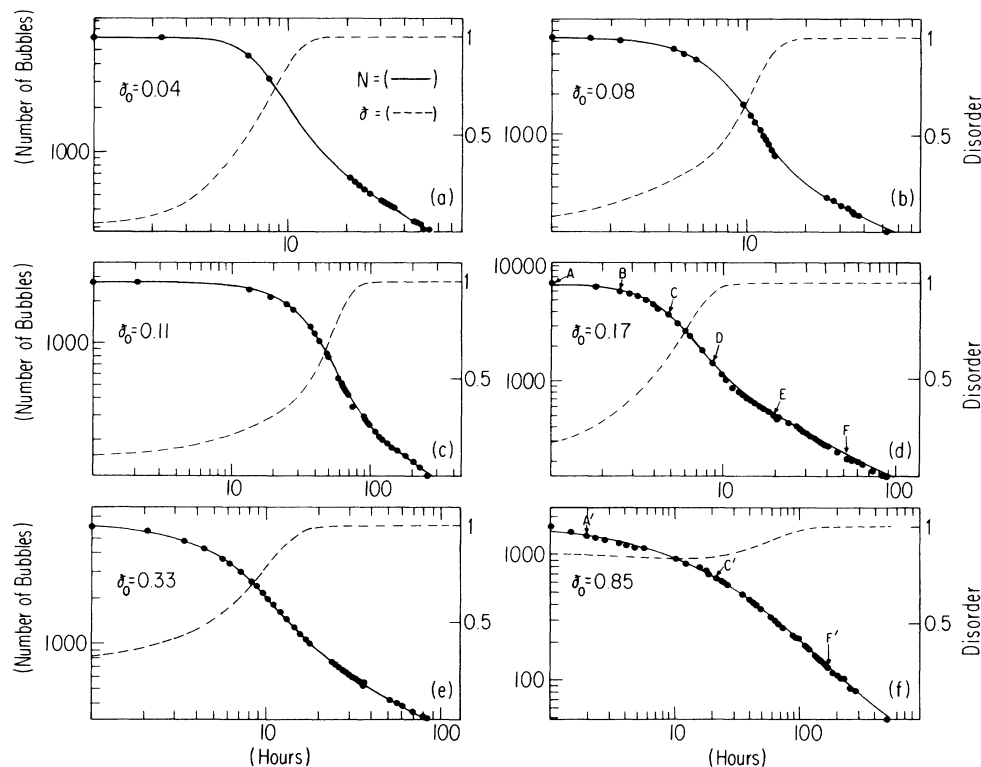For large initial disorder, the rate of evolution increases

FIG. 1. Number of bubbles and disorder $\partial(t)$ vs time for increasingly disordered initial conditions: (a) $\partial_0 = 0.04 \pm 0.02$, $\beta = 2.58 \pm 0.08$ (helium $\frac{1}{16}$ in. cell); (b) $\partial_0 = 0.08 \pm 0.01$, $\beta = 3.01 \pm 0.03$ (helium $\frac{1}{8}$ in. cell); (c) $\partial_0 = 0.11 \pm 0.02$, $\beta = 2.99 \pm 0.16$ (air $\frac{1}{8}$ in. cell); (d) $\partial_0 = 0.17 \pm 0.04$, $\beta = 2.48 \pm 0.05$ (helium $\frac{1}{8}$ in. cell); (e) $\partial_0 = 0.33 \pm 0.01$, $\beta = 2.89 \pm 0.04$ (helium $\frac{1}{16}$ in. cell); and (f) $\partial_0 = 0.85 \pm 0.05$, $\beta = 2.24 \pm 0.12$ (air $\frac{1}{8}$ in. cell); where $\partial_0 \equiv \partial(0)$. Errors are at 90% certainty. Capital letters in (d) and (f) indicate times referred to in the text and in Fig. 2. Dots are experimental values. Solid lines and values of β are best fits computed from the model. Dashed lines are the disorder as calculated from the model. Initial times are offset to 1 h.
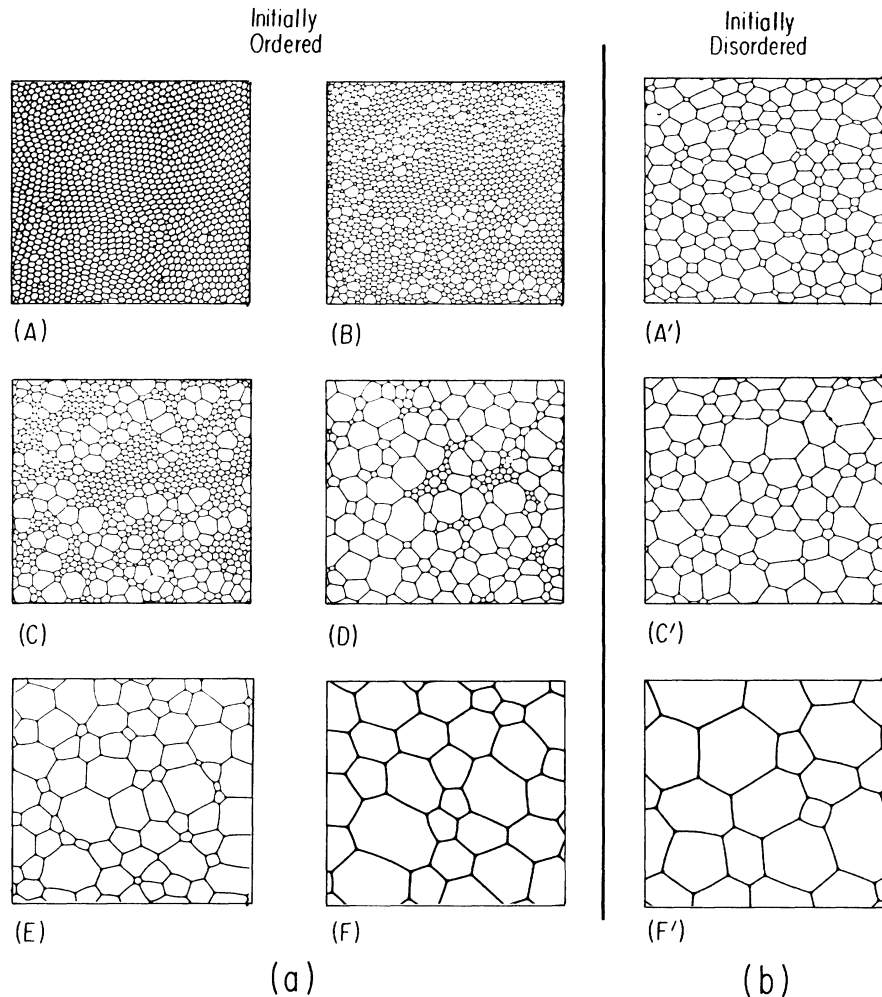
Initially
Ordered

Initially
Disordered

(A)   (B)   (A')

(C)   (D)   (C')

(E)   (F)   (F')

(a)   (b)

FIG. 2. Detail photographs of (a) an initially ordered run, see Fig. 1(d) (initial disorder $\vartheta_0 = 0.17$). Times are (A) $t = 1$ h, (B) $t = 2.52$ h, (C) $t = 4.82$ h, (D) $t = 8.63$ h, (E) $t = 19.87$ h, and (F) $t = 52.33$ h. (b) An initially disordered run, see Fig. 1(f) (initial disorder $\vartheta_0 = 0.85$). Times are (A') $t = 1.95$ h, (C') $t = 21.50$ h, and (F') $t = 166.15$ h. Capital letters are keyed to Fig. 1 and the text.

monotonically to its final value without overshoot. We present detail photos of such a run in Fig. 2(b) and the corresponding counts in Fig. 1(f). The labeled letters correspond to the time domains discussed above.

While we had expected that finite-size effects would be important in the latter stages of pattern evolution, the range of rollover points observed [ranging from 1000 bubbles for runs (a) and (d) to 100 bubbles in run (c)] suggests that the rollover is not an edge effect. To further control for edge effects we counted the number of bubbles touching the lateral walls of the cell (edge bubbles) as a function of time. If the average area of a bubble in contact with the edge is a constant times the average area of a bubble in the bulk, we would expect $N_{edge} \propto N_{bulk}^{0.5}$. This would be the case if the edge behaved as a noninteracting window on an infinite cell or as an infinite network of hexagonal bubbles. We find that $N_{edge} \propto N_{bulk}^{0.56 \pm 0.14}$ which is consistent with either hypothesis.

Previous authors have claimed that only three-sided bubbles can disappear directly and that four- and five-

sided bubbles must lose sides as they shrink.[1] However, we observe that both four- and five-sided bubbles can disappear directly, the ratio of direct disappearance to side shedding being nearly 1 for four-sided bubbles and approximately 0.1 for five-sided bubbles. This result supports recent theoretical work on this question.[10]

Many theoretical studies of soap bubble froths have assumed that the dominant mechanism for side redistribution is the pairwise exchange of sides among four vertices (the $T1$ process).[1] We find this is true only at short times when there are large residual stresses present from the filling. However, these stresses dissipate rapidly and after the first five minutes less than 1% of side redistribution is due to $T1$ processes unrelated to the disappearance of bubbles.

IV. VON NEUMANN'S LAW AND LEWIS'S HYPOTHESIS

Von Neumann proposed that the evolution of a two-dimensional soap froth could be modeled as a pure

diffusive process depending only on the geometrical structure of the bubble lattice.[7] He assumed that the gas was incompressible, so the rate of area transfer across a wall between two bubbles is simply proportional to the product of the wall area and the pressure difference between the bubbles. This pressure difference is, in turn, proportional to the curvature of the wall. Assuming that bubble walls are of constant curvature and thickness and that all walls meet in 120° angles, he obtained the following.

(i) The rate of growth for all n-sided bubbles is the same at a given time.

(ii) At a fixed time $\langle da_n/dt \rangle = \kappa(t)(6-n)$.

(iii) Rates are time independent, i.e., that $\kappa(t)=\kappa$ at all times.

We have evaluated $\langle da_n/dt \rangle$ at three stages of evolution corresponding to ordered, early disordered, and long-term disordered states.[3] We find that, at any given time, different n-sided bubbles evolve at different rates contradicting rule (i). We present the results for $\langle da_n/dt \rangle$ taken from the helium run shown in Fig. 1(d), in Fig. 3. The error bars show the spread in observed $da_{n,i}/dt$. Linear regressions on these data yield a value of $\kappa(t)=4.57\times10^{-2}\pm3.8\times10^{-3}$ mm^2/min for all three times. We are not interested in the actual value of κ, but in its constancy, which is better than 4%.

It is not clear why rule (i) should fail, since the derivation of Von Neumann's law is local. Spatial variations in system parameters, such as local film thickness or vertex angle, could affect local rates of diffusion. We have not attempted to measure these parameters, however. Somewhat surprisingly, rules (ii) and (iii) hold. While several factors would be expected to contribute to a time dependence in κ and hence in the rate of evolution, they do not seem to be important in practice.[12] In particular, if edge effects were important we would expect to see a change in the value of κ at long times.
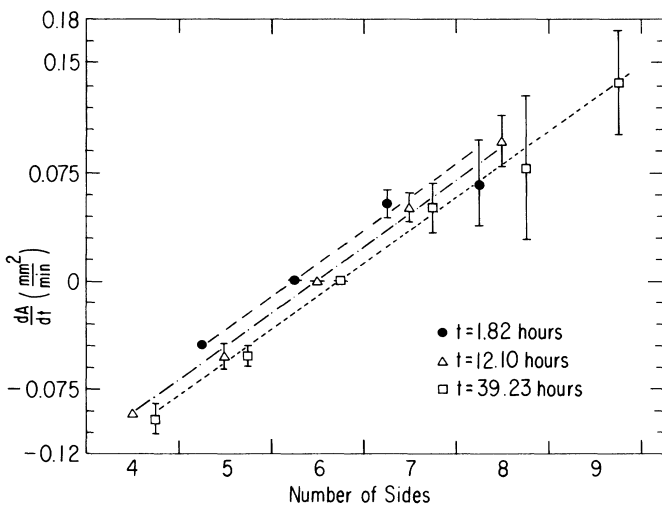
Since film creep and pressure equilibration times are very short compared to the typical diffusion time, κ is the only constant determining the rate of evolution. Thus the basic time scale in the experiment is constant to better than 4%. However, we cannot predict a priori the effective time scale of the redistribution of sides during the initial transient and this results in an uncertainty in the effective κ.

Lewis's hypothesis states that $\langle a_n \rangle = c_1+c_2n$ at any fixed time, where c_1 and c_2 are undetermined constants.[3,9] We estimate areas by connecting the vertices and centers of sides of bubbles by straight lines and measuring the area of the resulting polygon using a digitizing tablet. Since few-sided bubbles are convex and many-sided bubbles concave, we systematically underestimate the area of few-sided bubbles and overestimate the area of many-sided bubbles by up to a few percent.

We present experimental measurements of normalized bubble areas (i.e., $\lambda_n \equiv \langle a_n \rangle/\langle a \rangle$) as a function of n in Fig. 4. In spite of the bias noted above, we observe at all times that the area for few-sided ($n=3,4$) bubbles is larger than that predicted by Lewis's hypothesis. Indeed, for many runs, a linear fit actually predicts negative areas for three- and four-sided bubbles. Though the number of many-sided bubbles is too few to permit definite conclusions, the many-sided ($n>8$) bubbles in Figs. 4(b) and
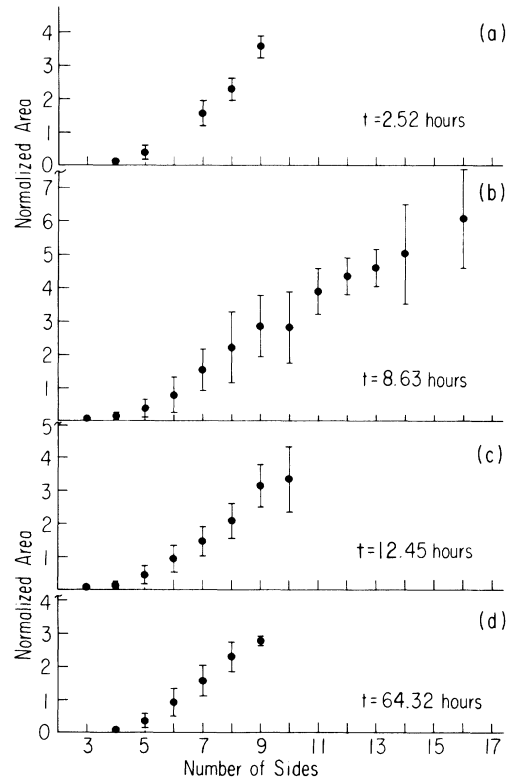

FIG. 3. Von Neumann's law, at $t=1.82$ h, $t=12.10$ h, and $t=39.23$ h, for the run given in Fig. 1(d). The measured $\kappa=4.57\times10^{-2}\pm3.8\times10^{-3}$ mm^2/min at all times. Error bars indicate the variation in κ among individual n-sided bubbles at 95% certainty.

FIG. 4. Normalized area vs number of sides $[\lambda_n(t)]$ at (a) $t=2.52$ h, (b) $t=8.63$ h, (c) $t=12.45$ h, and (d) $t=64.32$ h. All values are normalized by dividing by the average bubble area at the time of measurement. Error bars are 1 standard deviation and indicate the natural spread in distribution.

4(c) appear to be smaller than predicted. This discrepancy may be due to memory of the initial length scale. Statistical limitations prevent us from determining any more complicated law for the relation of area to number of sides. Significantly, the distributions of normalized area are constant within experimental errors (typically 5%) at all times. There is no sign of relaxation to an equilibrium state. We note that theoretical work by Rivier indicates that, by itself, a violation of Lewis's hypothesis implies that the distribution functions are not in statistical equilibrium.[13]

V. MODEL

Based on the above observations we propose the following phenomenological model for the transient. We divide the population into two classes, the bubbles in ordered regions and the bubbles in disordered regions, and denote the number of bubbles in each class by $O(t)$ and $D(t)$, respectively, with the total number of bubbles $N(t) \equiv O(t) + D(t)$. We assume that ordered regions do not evolve but are converted to disordered regions at a rate proportional to the contact area between order and disorder (that is, we make use of the observation that order is stable except where it is eaten away by disorder at the edges). To lowest order, assuming random distributions, the contact area is proportional to $O(t)D(t)/[O(t)+D(t)]$ which implies

$$\frac{dO(t)}{dt} = -\kappa_1 \frac{O(t)D(t)}{O(t)+D(t)} \ , \tag{1}$$

where κ_1 is a constant to be determined.

We also assume that the rate of disappearance of bubbles in disordered regions is independent of $O(t)$. Von Neumann's law (see Appendix) leads us to expect a power-law dependence $N(t) \propto t^{-\alpha}$ which is what we observe experimentally at long times when the system appears completely disordered. We therefore write an equation for $D(t)$ including terms for the conversion of order to disorder and for power-law dissipation,

$$\frac{dD(t)}{dt} = \kappa_1 \frac{O(t)D(t)}{O(t)+D(t)} - \kappa_2 D^\beta(t) \ , \tag{2}$$

where $\alpha = 1/(\beta - 1)$. Both $O(t)$ and $D(t)$ are abstract quantities. We have not attempted to put them in quantitative correspondence with the observed disorder. We can now define the abstract disorder

$$\vartheta(t) \equiv \frac{D(t)}{O(t)+D(t)} \ . \tag{3}$$

The parameter $\vartheta(t)$ gives information about the transition from order to disorder and has the practical advantage that it can be computed without calculating the distribution functions. While we can determine $\vartheta(t)$ directly from the data, we find it more convenient to fit $N(0)$, β, κ_1, κ_2, and $\vartheta_0 \equiv \vartheta(0)$ to give a minimal least-squares error against the experimental $N(t)$.[14] Values for $\vartheta(t)$ are shown in Fig. 1. For initially ordered runs, Figs. 1(a)–1(e), $\vartheta(t)$ increases smoothly to 1, reaching its final value where the experimental value of $N(t)$ rolls over into a power law. For the initially disordered run shown in

Fig. 1(f), the rate of conversion from order to disorder is slower than the rate of loss of disorder and $\vartheta(t)$ decreases slightly before increasing to 1 at the rollover. As expected, $\vartheta(t)$ is small for apparently well-ordered conditions, e.g., $\rho(5)/\rho(6) < 0.1$, and large for disordered conditions, e.g., $\rho(5)/\rho(6) > 1.0$. We obtain similar qualitative correspondence between $\vartheta(t)$ and other measures of disorder based on the distribution functions.

Results for the fits for $N(t)$ are also presented in Fig. 1. We obtain $\beta = 2.7 \pm 0.3$ corresponding to $\alpha = 0.59^{+0.11}_{-0.09}$. The typical error of the fits is better than 3%. The maximum observed error is 5%. In our two best runs, Figs. 1(d) and 1(f), the power-law behavior holds over a full decade. While we obtain consistent results for initial conditions ranging between 8000 and 1500 bubbles, we cannot rule out the possibility that other behaviors would be observed in much larger systems. We have tried, without success, a variety of other models based on chemical equilibrium equations and the master equation. None yield the desired exponent, though the master equation does reproduce certain features of the evolution of the distribution functions. Marder, using a Von Neumann's law based mean-field model, obtains excellent qualitative agreement with the experimental results. However, while he observes the two regimes of exponential and power-law growth, he obtains $\alpha = 1$ and different values for the λ_n's.[15]

VI. CONCLUSIONS

We conclude that the two-dimensional soap bubble lattice exhibits two distinct states: a transient characterized by the presence of crystalline order and a long-term fully disordered state characterized by a power-law growth in bubble area, $\alpha = 0.59^{+0.11}_{-0.09}$. Smith and Weaire obtained an incorrect value for this exponent because both mistook the high-evolution rate transient for limiting behavior. Previous authors have interpreted the broad area distribution of the transient as indicating a fractal structure;[5,6] however, our observations of the distributions of bubble areas and number of sides rule out such a structure for the developed bubble lattice. We have not, however, calculated higher moments of the distribution functions. Apparently disordered states can contain residual order, i.e., have $\vartheta(t) < 1$, as seen in Fig. 1(f). The disorder produced by an irregular filling process is not the same as that produced by natural relaxation nor do initially disordered systems necessarily reach a long-term state faster than initially ordered systems.

We have experimentally verified the statistical form of Von Neumann's law to 4%. This law poses strong constraints on the dynamics of the lattice. It implies that there is no nontrivial equilibrium. More generally we have argued on dimensional grounds that if κ is constant Von Neumann's law implies $\langle a \rangle \propto t$. We observe no sign of such behavior in any of our experimental runs. Neither do we see any tendency for $d\log\langle a \rangle/d\log(t)$ to increase in time, which would indicate a gradual drift towards such an equilibrium. There must be a hidden time scale in the evolution. We conclude that there are no long-term equilibrium states except for the perfect hexagonal lattice

and the empty lattice. The origin of the anomalous exponent α remains an open question.

ACKNOWLEDGMENTS

APPENDIX: CALCULATIONS OF LONG-TERM BEHAVIOR FROM VON NEUMANN'S LAW

We wish to calculate the behavior of the system assuming that it has reached an equilibrium, i.e., scaling state. We present two arguments, one dimensional, the other explicitly based on Von Neumann's law.

We may argue dimensionally as follows.[16] We have asserted that the only significant parameters for long-term states, i.e., states which are independent of initial conditions, are κ, $\langle a \rangle$, and t. We have checked this experimentally by observing that behavior is independent of cell size, cell thickness, and the diffusing gas. The combination

$$\frac{1}{\kappa}\frac{d\langle a \rangle}{dt} = f(t, \langle a \rangle) \tag{A1}$$

is dimensionless. However, there is no way to form a dimensionless number using only t and $\langle a \rangle$, therefore the right-hand side must be constant, and we conclude that in equilibrium either $\langle a \rangle \propto t$ and $N \propto t^{-1}$, where N is the number of bubbles in a fixed area, or $\langle a \rangle = $ const and $N = $ const. The first case corresponds to $\beta = 2$ in the model, the second to an empty or pure hexagonal lattice. In general any power-law dependence of the various "constants" on N (e.g., of κ as experimentally observed) will add a correction term to the exponent α and hence also to

β in the model.

Alternatively, we may argue from Von Neumann's law as follows. Let $\rho(n)$ be the probability that a bubble selected at random has a given number of sides. Let $\langle a_n \rangle$ be the average area of an n-sided bubble in the interior of the cell. Let $N_0(t)$ be the number of bubbles in the interior. Let $A_0(t)$ be the area of the interior of the cell. Let A be the total area and define $N(t) \equiv N_0(t)A/A_0(t)$ to be the corrected bubble count. Let $N_n \equiv \rho(n)N$ be the corrected number of n-sided bubbles and $\lambda_n \equiv \langle a_n \rangle N(t)/A$. Then

$$\frac{d\langle a_n \rangle}{dt} = \frac{d}{dt}\left[\frac{1}{N_n}\sum_{i_n=1}^{N_n} a_{i_n}\right] , \tag{A2}$$

where i_n indexes all n-sided bubbles at time t. Taking the derivative and substituting Von Neumann's law we obtain

$$\frac{d\langle a_n \rangle}{dt} = -\frac{1}{N_n}\frac{dN_n}{dt}\langle a_n \rangle + \kappa(n-6) . \tag{A3}$$

If we assume that both λ_n and $\rho(n)$ are constant at equilibrium we obtain

$$\lambda_n\left[\frac{d\langle a \rangle}{dt} + \frac{1}{N}\frac{dN}{dt}\langle a \rangle\right] = \kappa(n-6) . \tag{A4}$$

But the left-hand side is 0 since

$$\langle a \rangle \frac{1}{N}\frac{dN}{dt} = -\frac{d\langle a \rangle}{dt} ,$$

so this situation is possible only in the trivial cases of the pure hexagonal and empty lattices. Thus there is no strict nontrivial equilibrium. If we allow variation of λ_n and $\rho(n)$ we obtain from Eq. (3) the conditions

$$\frac{1}{\rho_n}\frac{d\rho_n}{dt}\lambda_n + \frac{d\lambda_n}{dt} = \frac{\kappa(n-6)}{A}N . \tag{A5}$$

As noted previously, we observe experimentally that λ_n is constant.

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[1]D. Weaire and N. Rivier, Contemp. Phys. **25**, 59 (1984). This is the standard review paper on soap froths.

[2]C. S. Smith, *Metal Interfaces* (American Society for Metals, Cleveland, 1952), p. 65.

[3]Unsubscripted averages are taken over all bubbles in the selected area, averages with a subscript n (e.g., $\langle a_n \rangle$) are taken over all n-sided bubbles. However, the average $\langle da_n/dt \rangle$ is taken only over bubbles which remain n sided over the δt (typically $\frac{1}{2}$ h) used to define the derivative.

[4]D. A. Aboav, Metallography **13**, 43 (1980).

[5]For a clear exposition of this controversy see D. Weaire and J. P. Kermode, Philos. Mag. B **47**, L29 (1983).

[6]D. Weaire and J. Wejchert, *Computer Simulation of Microstructural Evolution* (The Metallurgical Society, Warrendale,

1986), p. 49.

[7]J. Von Neumann, *Metal Interfaces* (American Society for Metals, Cleveland, 1952), p. 108.

[8]D. Weaire and T. Fu of Trinity College, Dublin are currently studying Von Neumann's law using a slightly different technique from that described in this paper.

[9]F. T. Lewis, Anat. Rec. **38**, 341 (1928).

[10]D. Weaire (unpublished).

[11]C. Beenakker, Phys. Rev. Lett. **57**, 2454 (1986).

[12]Factors which could reduce diffusion rates and hence $\kappa(t)$ are (i) thickening of the soap films and Plateau borders as the total film length decreases (we work with a fixed volume of fluid), (ii) leakage of gas into the cell, and (iii) reductions in pressure differentials resulting from gradual equilibration. However, for short times and well-ordered states, the average pressure difference must increase with time.

[13]N. Rivier, Philos. Mag. B **47**, L45 (1983).

[14]We may reduce this worryingly large number of free parame-

ters as follows. We solve for $\partial(t)$ in terms of $N(t)$ and $dN(t)/dt$,

$$\partial(t)=(\kappa_2)^{-1/\beta}\frac{\left[-\dfrac{dN(t)}{dt}\right]^{1/\beta}}{N(t)} \ .$$

This equation determines ∂_0 locally from the experimental data. We determine β and κ_2 with reference to limiting behavior only, by defining

$$\beta\equiv\lim_{t\to\infty}\frac{d\log[N(t)]}{d\log(t)}$$

and

$$\kappa_2\equiv-\lim_{t\to\infty}\frac{dN(t)}{dt}N(t)^{-\beta} \ .$$

In this way we reduce the number of global fitting parameters to one, κ_1. Fits performed using this technique are compatible with the results presented in Fig. 1. However, because of the limited number of decades of data, we obtain better accuracy doing a full five-parameter fit.

[15]M. Marder, Phys. Rev. A **36**, 438 (1987).

[16]This argument was suggested by Professor Bernard Castaing, currently at the James Franck Institute, University of Chicago, Chicago, Illinois.