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Evolution of grain boundaries in two-dimensional foam

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Abstract. The temporal evolution of disorder around grain boundaries between domains of ideally six-fold coordinated two-dimensional foam has been studied experimentally, using a foam comprising bubbles bridging between a soap solution and a cover glass. The disorder, quantified by the second central moment of the distribution of topological classes of the cells (μ_2), generally increases. In certain cases, in which the evolution can be followed over longer times, μ_2 eventually falls. This may be connected with the transient peaks for μ_2 found in previous studies of relatively ordered soap froths. The absolute values of μ_2 depend upon the boundary conditions imposed upon the foam, a rigid wall leading to higher values than a deformable boundary. The disorder about the grain boundaries propagates into the adjacent regions of ordered foam with constant speed, the roughness of the interface increasing with time.

1. Introduction

There has been a resurgence of interest in the physics of foam, particularly as a model of disordered systems [1, 2]. While the situation is changing as the structure and dynamics of three-dimensional foam becomes more accessible to various probes [3–5], and computers become more powerful, there has been a historical tendency to study the 2D case as more accessible to both experimental and computational investigation. Much interest has focused on the temporal evolution of 2D foam as it coarsens. This evolution in 2D proceeds via various basic topological processes [1], of which neighbourhood switching (T_1) and the disappearance of small (usually three-sided) cells (T_2) are the only ones of present concern. Coarsening in 2D foam as described by von Neumann's law,

$$\frac{\mathrm{d}A}{\mathrm{d}t} = k(n-6) \tag{1}$$

is due to the diffusive exchange of gas from small to large cells. Here n is the number of neighbours (n is also referred to as the topological class of the cell) and A the area of a given cell. Here we focus on the evolution in one specific situation: foam containing a grain boundary between two areas of ideal foam.

In its temporal evolution 2D foam has been found to reach a stationary scaling state at long times in which the topological disorder quantified by μ_2, the second central moment of the distribution of n, becomes independent of the initial state of the system [6]. While an initially disordered 2D foam makes a smooth transition to this final state, μ_2 for a foam which is initially

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rather ordered passes through a transient peak before reaching the apparently universal limiting value. The origin of this initial transient has been a matter of some recent interest: in a 2D foam with a small but non-vanishing degree of disorder most cells will have $n = 6$, but for non-zero μ_2 there must be defects in the system, for which $n \neq 6$. These will comprise point defects (dislocations and bubbles large or small enough that they are not hexagonally coordinated) and larger areas of disorder, including grain boundaries. Various recent simulations [7–9] and experiments [10, 11] have concentrated on the evolution of disorder around single point defects in otherwise ideally six-fold coordinated foams. It is found that for such isolated defects μ_2 of the area of disorder about the original defect increases monotonically with time, never exhibiting the peak seen in the early evolution of the relatively ordered foam [6]. Certain maxima found experimentally appear attributable to artefacts. However, when two or more spatially separated dislocations are introduced into an ideal foam, μ_2 does seem to show a real maximum in its temporal evolution [11], arising from the interaction of the growing areas of disorder about the separate initial defects. There is, therefore, some interest in the evolution of grain boundaries, which present a linear array of spatially separated dislocations (each comprising a bound pair of five- and seven-coordinated cells).

There are only limited data on this point: brief references in experimental and simulation studies state that the area of disorder along the grain boundary spreads into the ordered regions of foam, a gradient of cell size developing normal to the line of the boundary [6, 12]. Some data of rather limited precision have been reported [13] for a boundary between two ordered domains in a model 2D foam resembling that used in the present work. We present more comprehensive data from a series of experiments on the evolution of various types of grain boundary in otherwise ideal two-dimensional foam. We concentrate on such issues as the increase of disorder in foam around the different types of boundary, the effect of different boundary conditions imposed upon the foam and of the angular mis-orientation of the domains of ideal foam adjacent to the boundaries. We also consider the propagation of the disordered region into the ordered foam, leading us to investigate the width and roughness of the interfaces.

Our model 2D foam comprises bubbles bridging between the surface of a soap solution and a glass cover plate a few mm above the solution. This cover glass prevents loss of gas from the bubbles to the atmosphere so that the raft of bubbles has an essentially indefinite life, allowing its evolution to be studied over extended times. The sessile bubbles are flattened by buoyancy, causing the raft to be of almost constant thickness, making it a good model of a 2D foam. No bubbles ever move into the third dimension during our experiments.

While there have been previous studies of grain boundaries in bubble rafts [14], these have had a very different focus from the present work. They entirely concerned the modelling of grain boundaries in materials, building on the pioneering use of the bubble raft as a 2D crystal analogue [15], rather than the present concern with the evolution of the bubbles *per se*.

2. Methods

We have studied three types of grain boundary: one (referred to as a grain boundary loop) surrounding a lattice-like domain mis-orientated with respect to the foam occupying the rest of the cell, the second comprising a quasi-linear boundary between two areas of foam having different bubble sizes (‘incommensurate grain boundary’) and finally relatively low angle grain boundaries between areas of ideal foam. Here we describe the creation of these grain boundaries.

The first two types were made in the same cell as used in our previous studies [10, 11, 16]: a metal plate some 1 cm thick and containing a hexagonal hole 6 cm on a side was placed in a soap solution so that its upper surface was 2–3 mm above the liquid, and covered with a glass

plate. N_2 gas was then bubbled into the cell through a long hypodermic needle; by sweeping the needle to and fro ideally hexagonal foam could be generated. Any dislocations occurring in otherwise ideal foam could be removed by manipulation [11]. The cell was placed upon a light box to allow photography of the foam at various stages of its evolution.

To make a grain boundary loop, a belt of ideal foam was formed around the cell walls, some five to seven bubbles wide. An isolated patch of ideal foam was then generated in the centre of the cell. As more bubbles were added, this patch spread towards the foam lining the walls. Bubbling N_2 was stopped just as the two areas of foam came into contact.

Due to the symmetry of the hexagonal cell it was not possible to form separate mis-orientated areas of ideal foam. Apart from the loops just described, the only grain boundary which could be formed was thus between foams comprising different size bubbles. The cell was first approximately half filled with uniform foam of bubbles of one size, then the rest was filled with bubbles of a different size using a hypodermic needle of different gauge. The number of dislocations in the boundary increased with the size difference between the bubbles; a very small difference led to a curved interface rather than a line of dislocations.

Creation of a grain boundary between mis-orientated areas of ideal foam requires a modified approach. A cell is needed in the form of a distorted hexagon, each half being a quadrilateral with base angle 120°, these parts being slightly mis-orientated with respect to each other (figure 1). The cell comprised a Perspex base in which were set six brass pegs round which a large rubber band could be stretched to define the area of the foam. Three of the pegs could be moved to yield different degrees of mis-orientation (0, 4.5 and 8.9°). The method of forming grain boundaries in this cell was as described above for the incommensurate case.

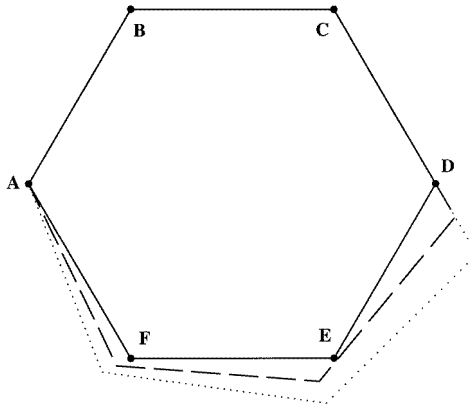

Figure 1. A diagram of the cell for creation of low angle grain boundaries. Of the six points indicated, three (D–F) can be moved so that the two halves of the cell make an angle of 8.9° (dotted line), 4.5° (dashed line) or zero (full line).

Unlike the previous cell, in which the foam encounters a rigid boundary, the boundary of this new cell was deformable. We believe that the different boundary conditions (referred to below as 'hard' and 'soft') imposed by the two cells underlie certain differences in the behaviour of the foam to be described below.

3. Results and discussion

We first present data concerning the growth of disorder around the initial grain boundaries. We subsequently turn to the roughening of the boundaries.

3.1. Topological disorder

As is conventional, we define the degree of disorder via the second central moment (μ_2) of the distribution of the numbers of neighbours ($P(n)$) of the bubbles in a 'cluster' of bubbles about the original defect [7, 8, 10, 11]. Here we consider the line of dislocations forming the grain boundary as a single defect. The cluster is defined as that set of bubbles around the dislocations which have at least one neighbour which is not six-fold coordinated, plus the belt of six-coordinated bubbles separating the dislocations along the line. This definition is somewhat arbitrary (see [8] for an alternative definition for an isolated defect), and consequently the absolute values of $P(6)$ and μ_2 reported here are not very significant in themselves. We thus focus upon the temporal changes of these statistics.

Increasing disorder first appeared around the original dislocations defining the grain boundary some 10–13 hours after formation of the foam and grew due to coarsening (figure 2). The disorder propagated outwards in time, as for point defects [7, 10, 11]. As the grain boundary evolved a gradient of cell size developed in the direction of spreading. The bubbles were generally largest along the original line of dislocations, although due to the wetness of our foam (which leads to a lack of T_2 processes [10, 11]) a number of small bubbles persisted adjacent to the largest bubbles (see figure 2). This general behaviour accords with that found in simulations [12], and in earlier experiments [6, 13].

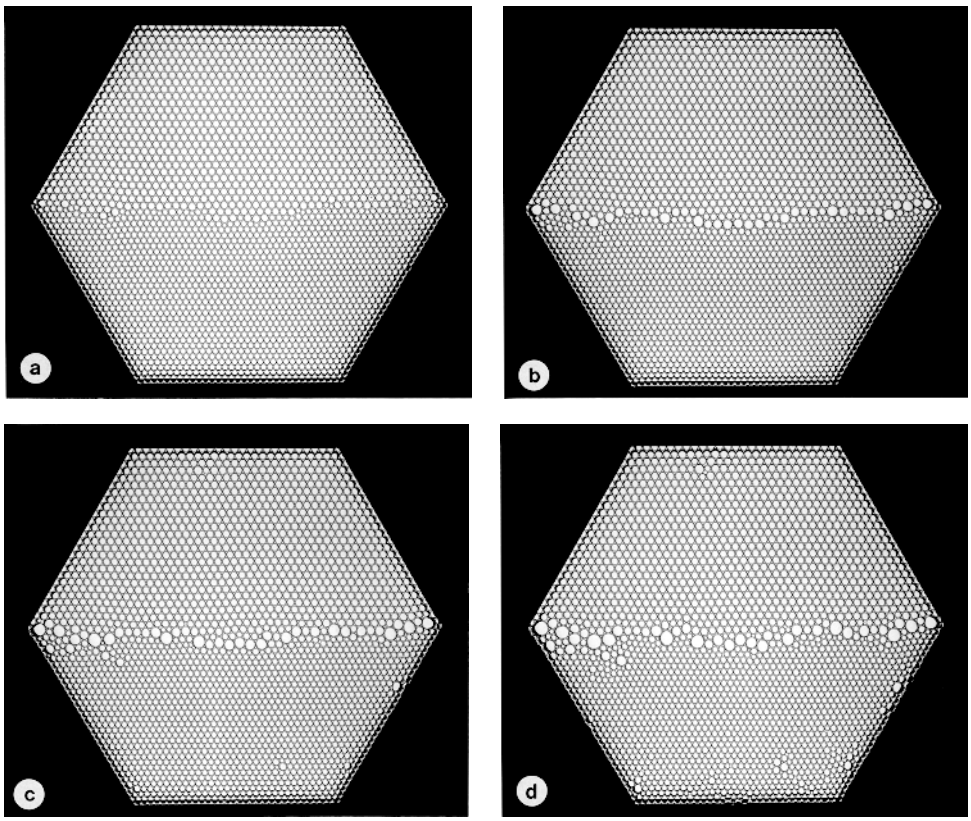

Figure 2. Pictures of a typical evolving foam having an incommensurate grain boundary: (a) as formed, containing 16 dislocations ($t = 8$ h); (b)–(d) after (b) $t = 15$, (c) $t = 26$ and (d) $t = 35$ h.

The time over which we could follow the growth of disorder was limited by the appearance of generalized disorder in different areas of the foam, over time scales of the order of days (evident in figure 2(d)), due to coarsening of the 2D foam arising from inevitable tiny differences in the size of the 'ordered' bubbles in the body of the foam. Eventually the cluster grew into this coarsening-induced disorder; all the data presented here relate to times before this occurs.

As in previous studies [10, 11], we use the number of bubbles in the cluster (n_c) as the independent variable, instead of time. This helps to obviate certain empirical fluctuations. (n_c varied relatively smoothly with time in all experiments.) The actual rate of growth of the cluster differs from case to case. For example, for both incommensurate and loop grain boundaries the rate of evolution of the cluster could be varied experimentally. By inserting more bubbles in one area of the foam (creating more dislocations) the foam was made more compact, so that the liquid films between the bubbles were thinner, allowing more rapid evolution and the faster appearance of disorder.

3.1.1. Incommensurate grain boundaries. Figure 2 shows a typical example of the evolution of an incommensurate grain boundary. After 8–10 hours, during which time no great change was observed, disorder increased around the initial dislocations. Figure 3 shows the distribution of the topological classes of the bubbles in the cluster and its evolution for a rather compact foam. Initially all the bubbles in the cluster, except the five- and seven-coordinated ones defining the dislocations, have $n = 6$, but with time $P(6)$ falls and $P(n)$ extends to larger and smaller n; the peak of the distribution, however, remains at $n = 6$. The general features of these $P(n)$ resemble those observed for foam evolving about point defects [10, 11]. However, they differ from those for many 2D cellular networks: in particular $P(6)$ is always relatively high and we observe a significant population at $n = 3$. These three-coordinated bubbles are very small, and tend to lie around large bubbles. The disappearance of these small bubbles via the T_2 process is somewhat inhibited by the slow diffusion of N_2 from them to their neighbours due to the small area of contact [10, 11].

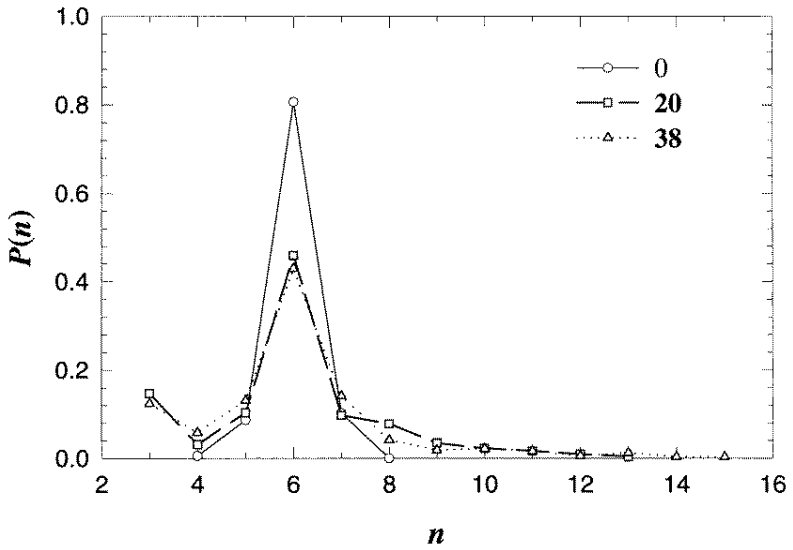

Figure 3. Topological class distributions for foam containing an incommensurate grain boundary. Times are as in the legend.

The increase in disorder reflected by the widening of $P(n)$ can be characterized through the temporal evolution of μ_2, shown in figure 4 for three grain boundaries with different initial numbers of dislocations. μ_2 increases to rather high values, but it ultimately decreases. The larger the initial number of dislocations, the higher the peak value of μ_2. This does not appear to be a very fundamental property, but rather results from the arbitrary nature of the cluster: for fewer dislocations in the grain boundary the belt of six-coordinated bubbles makes a larger contribution to the initial $P(n)$, reducing the values of μ_2. Bubbles in this belt do not evolve to such levels of disorder as those about the dislocations.

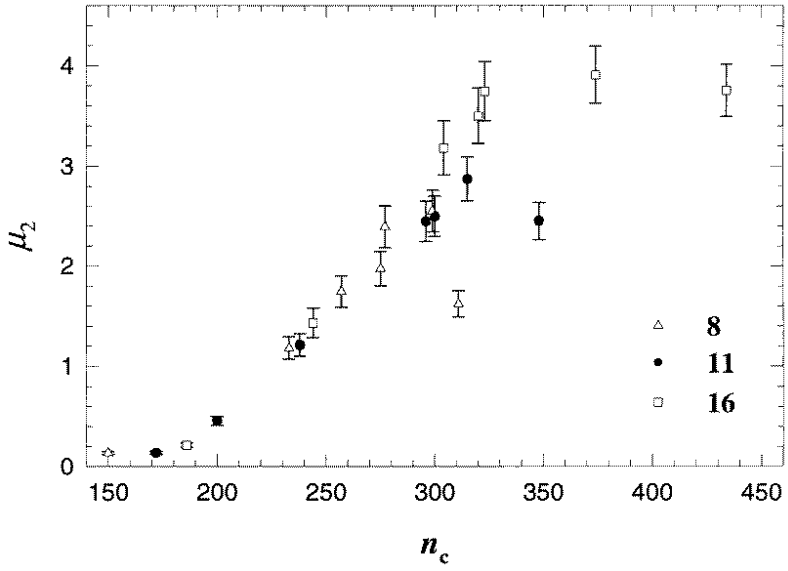

Figure 4. The evolution of μ_2 with n_c for foams with incommensurate grain boundaries having different initial numbers of dislocations (see the legend). See the text for a discussion.

It has recently been argued [13] that peaks in μ_2 such as those seen in figure 4 (and, by implication, that seen as relatively ordered foam evolves to the scaling state [6]) arise from the interaction of growing clusters of disorder about separated dislocations. In fact for these grain boundaries the observed peaks occur long after the time when the regions of disorder growing about individual dislocations impinge upon each other. To what can the eventual decrease be ascribed? For single isolated defects such maxima seem to be artefacts of the wetness of the foam, arising due to the ultimate disappearance of the small three-coordinated bubbles. However, as for spatially separated dislocations [11], this does not seem to be the case for these grain boundaries; indeed the absolute number of such bubbles grows in some cases, those which disappear being replaced from the population of four- and five-coordinated bubbles. Rather $P(n)$ is decreased at its extremes, while the central peak for $5 \leqslant n \leqslant 8$ broadens as the foam coarsening leads to a more generally disordered state.

These observations are in general agreement with the growth of disorder about dislocations [11], both isolated and in groups (bound pairs or spatially separated). μ_2 attained a rather small value for a single dislocation, a larger one for a bound pair of dislocations and a still higher transitory peak value when two dislocations were spatially separated, the latter effect arising from the interaction between the growing clusters about the two dislocations. For the grain boundaries, having several separate dislocations, the peak value of μ_2 is comparable to or

higher than that found previously for spatially separated dislocations [11].

3.1.2. Grain boundary loops. Figure 5 shows a typical example of a foam containing a loop grain boundary and its evolution. As before, the peak of $P(n)$ (data not shown) was always at $n = 6$, the tail of the distribution extending to larger n as time progresses. In this case μ_2 increases monotonically (figure 6), reaching values comparable to those seen for incommensurate grain boundaries containing many dislocations, but not showing the eventual decline seen for that system. We believe that the latter difference arises from the experimental cut-off as the cluster evolves into regions of generalized disorder (figure 5(d)). For loop grain boundaries this happens relatively early because the loop is always rather close to the periphery of the cell, from where such external disorder frequently appears.

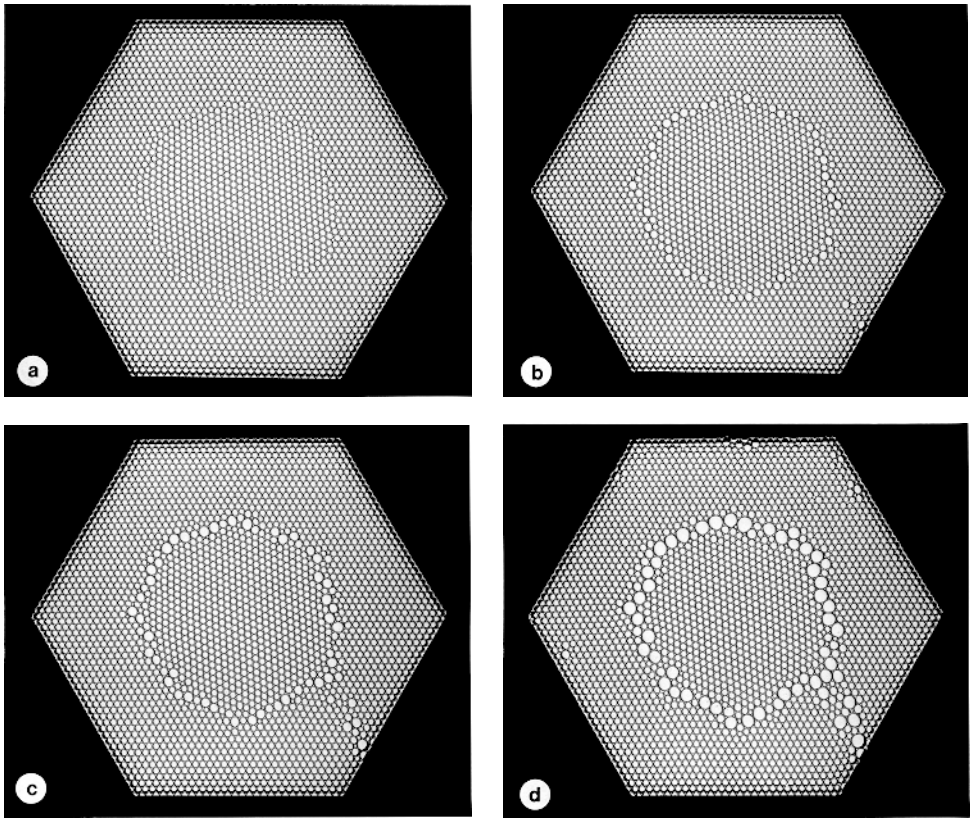

Figure 5. Images of a 2D foam containing a loop grain boundary (a) as formed and (b)–(d) after (b) 16, (c) 24 and (d) 36 h. Between (c) and (d) a region of generalized disorder has grown into the evolving cluster around the grain boundary, terminating the analysis.

A few general observations are relevant. The loop grain boundaries are generally polyhedral in shape, so that they comprise various lines of different mis-orientation between the inner and outer ordered regions of foam. It is significant that the growing disorder is very similar on all sides of the loop (e.g., figure 5(c)), indicating that the angle of mis-orientation does not really affect the evolution of disorder in the foam. Again, as mentioned in section 2, the bubbles inside and outside the boundary are similar in size. The similar general behaviour to that for the incommensurate grain boundaries discussed above suggests that the difference

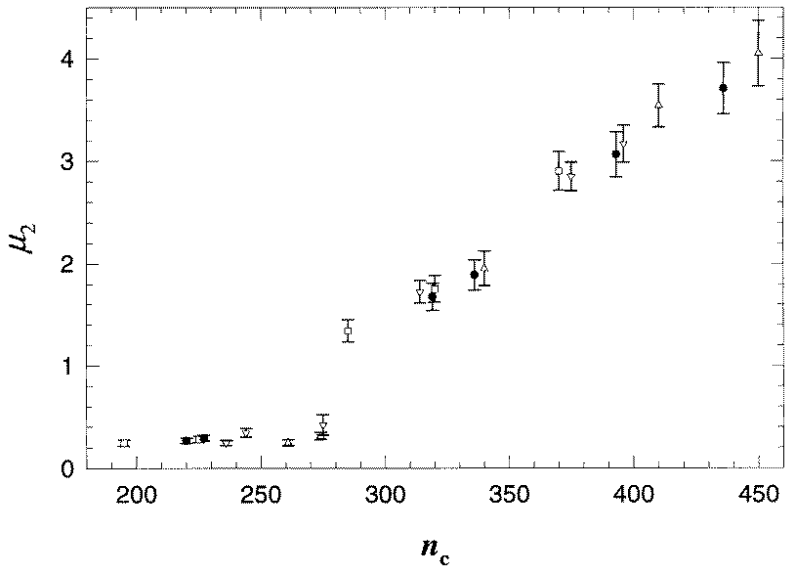

Figure 6. The temporal variation of μ_2 for three different foams containing loop grain boundaries. See the text for a discussion.

in bubble size in the latter does not affect the evolution of disorder very significantly. We conclude that the major influence leading to large absolute values of μ_2 is the presence of many dislocations along the grain boundary. This is not entirely surprising, as the cluster (as defined here) about a grain boundary with few dislocations will contain many 'spectator' six-coordinated bubbles in the belt joining the dislocations. As we have seen above such bubbles act to prevent μ_2 from rising too far by keeping $P(6)$ high.

3.1.3. Low angle grain boundaries. Figure 7 illustrates the evolution of a grain boundary between mis-orientated arrays of bubbles of different sizes. The degree of disorder is manifestly relatively low compared to that for the incommensurate grain boundaries described above (cf figure 2). We recall that the low angle grain boundaries are formed in a soft-sided cell, rather than the rigid cell used for the earlier types, and ascribe the differences in the evolution of disorder to this fact. Indeed, we observe similarly low degrees of disorder for foams in the soft cell even when the angle of mis-orientation is zero, leading to a grain boundary essentially just like the incommensurate cases treated above.

The topological class distributions (data not shown) confirmed this impression of relatively low disorder, $P(6)$ (a measure of the degree of order) always remaining rather high compared to those of figure 3. The evolution of μ_2 (figure 8) thus differed from the previous variations, the increase reaching only rather modest values. No terminal decrease of μ_2 is observed, and it may be that the observed values are tending to saturate (more apparent when plotted versus time, rather than n_c [17]).

The temporal variation of μ_2 does not depend to any great extent upon the mis-orientation involved (figure 8), confirming the inference drawn above from the loop grain boundaries that changes in this angle have little or no effect upon the growth of disorder about the boundary.

We attempted to study the evolution of a low angle grain boundary separating ordered areas of foam comprising equal-sized bubbles. Unfortunately there was no growth of disorder

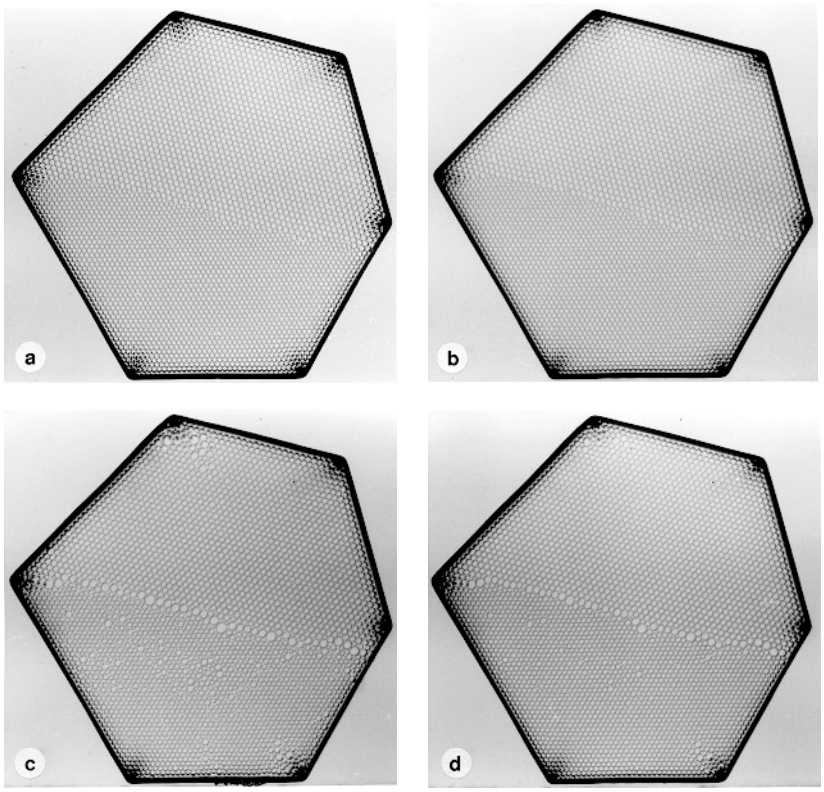

Figure 7. A typical foam with a low angle grain boundary between areas of ideal coordinated bubbles of different sizes for times (a) 0, (b) 22, (c) 34 and (d) 42 h.

about the grain boundary. Rather we observed an eventual relaxation of the entire system, in which cracks appeared at various points throughout the foam. The time scale for this to happen—over 24 h—was consistent with that for coarsening of the foam (i.e., the appearance of generalized disorder, as already discussed). We do not fully understand this phenomenon, but it clearly derives from the different boundary conditions imposed upon the foam in the soft cell, as the loop grain boundary involves equal-sized bubbles, but does display a more normal evolution pattern.

3.2. Comparison between rigid and soft cells

Why does foam reach a higher degree of disorder in the rigid cell than in the soft cell? As we have noted, differences in either the grain boundary angle or the bubble size cannot be responsible, as shown by the similarity of the values of μ_2 achieved in the loop and incommensurate grain boundaries. The difference must lie in the cells themselves. The cells were of different sizes, the soft one being some 10 cm on a side, whereas the rigid one was 6 cm. However, experiments with a 10 cm rigid cell showed exactly the behaviour shown here for the smaller cell. We are thus left with the different boundary conditions themselves.

Our foams are in a state of homogeneous tension due to inter-bubble attraction [16]; the walls exert a further attraction upon the nearest few layers of bubbles. Unlike the rigid walls of the first cell, the periphery of the soft cell, being made of an elastic material, can yield and de-

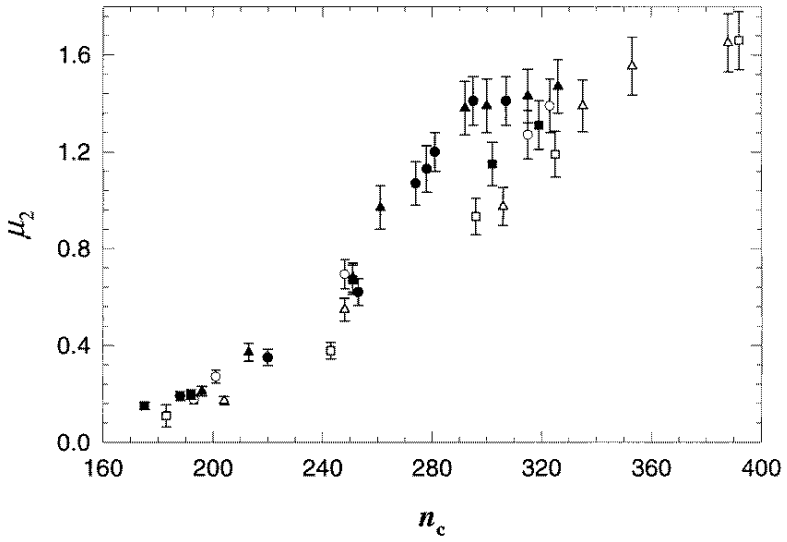

Figure 8. The evolution of μ_2 for six low angle grain boundaries. The various symbols indicate different values of angular mis-orientation 0° (○), 4.5° (□) and 8.9° (△). See the text for a discussion.

form in response to this tension. In particular, evolution of the foam (along the grain boundaries and, to a lesser extent, elsewhere in the foam) will modify this tension, as bubbles grow and shrink, and the soft periphery can adapt to such changes, effectively reducing the magnitude of the overall tension. This apparently somewhat inhibits the tendency to further evolution.

These differences between the two cases serve to point up the difficulty of forming a model which adequately mimics a real physical system. In real foam of any appreciable extent a given sub-set of bubbles would presumably correspond more closely to our 'soft' boundary conditions than to the 'hard' ones applicable to most of our work, as the surrounding foam could expand or contract in response to changes in the sample sub-set. Such an explanation probably explains why the maximal values of μ_2 found in a previous experimental study of the evolution of grain boundaries [13] were rather low ($\sim$1.3); the grain boundary studied in that work looks as if it spanned only a fraction of the entire foam.

These things are simpler for computer simulations, in which the use of periodic boundary conditions may remove such boundary effects. However, in the only computer simulation which addressed grain boundaries in foam [12], periodic boundary conditions were not imposed, but rather the foam was surrounded by a rigid boundary. The initial state comprised a continuous line of dislocations in a polydisperse array of hexagonal cells, introduced through a series of T_1 processes. While μ_2 referred to the entire foam, the disorder around the 'grain boundary' filled the foam sample in the final state, so that it may not be very incorrect to compare that value of μ_2 with the present data for the cluster of disorder about the incommensurate grain boundaries. The final value of μ_2 given by [12] is 3.24, which is clearly rather similar to the peak values found here for incommensurate grain boundaries comprising many dislocations. Too much weight should not be attached to this agreement, based as it is on a single simulation run. However, it does suggest that further simulations comparing the consequences of both rigid and periodic boundary conditions could be enlightening.

3.3. Roughening of grain boundaries

We turn to considerations of the propagation of the interfaces between the ordered foam domains and the region of disorder about the grain boundary. Many such propagating interfaces display fractal roughening [18]. We briefly outline current approaches to these matters. An interface can be reduced to the set $\{i\}(i \leqslant L)$ of points which lie furthest into the hitherto unperturbed medium (this removes any arbitrariness due to the interface folding back upon itself—cf the lower edge of the grain boundary in figure 2(c)). The height of the interface at time is defined as the average over i of the distance $h(i)$ of that point from its starting position: $\langle h(t)\rangle$. For a constant rate of propagation of the disturbance generating the interface $\langle h(t)\rangle \propto t$. The roughening is quantified via the width of the interface, defined as the standard deviation $w(L,t) = \sqrt{\langle [h(i,t) - \langle h(t)]^2\rangle}$.

In the present case the area of disorder grows into two adjacent homogeneous media, and so we define h as the transverse distance between the two bubbles adjoining opposite sides of the cluster. The number of such bubbles along the interface across the cell is taken as L. For linear grain boundaries in both hard and soft cells we find that $\langle h(t)\rangle$ increases linearly with t (figure 9), although the absolute values differ in the two cases, as is apparent from figures 2 and 7. (In this case we use real time rather than n_c.) Thus the disorder about the grain boundaries propagates into the ordered foam with constant velocity, as is commonly found for many other instances of interfacial growth [18].

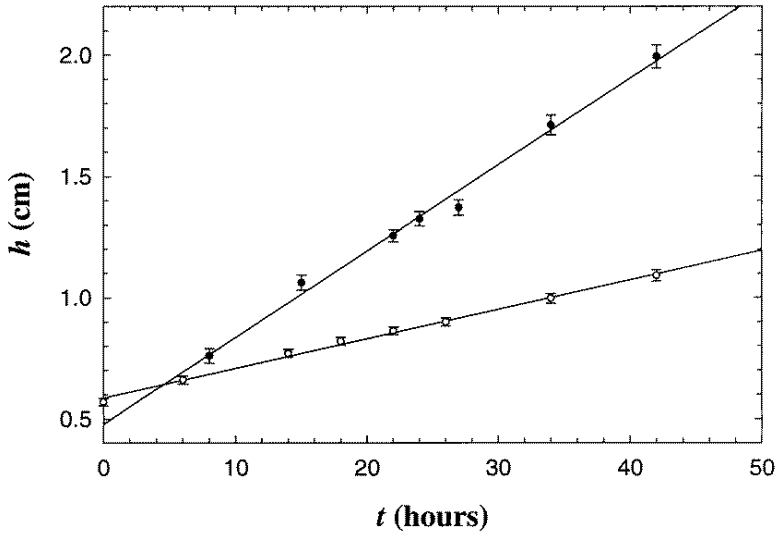

Figure 9. The variation of the height $\langle h(t)\rangle$ with time for an incommensurate grain boundary (●) and for a low angle grain boundary (○).

The roughness $w(L,t)$ only increases after an initial transient decrease (figure 10). The latter is a consequence of the discrete cellular nature of our medium. $h(i, t = 0)$ for the initial interface (as defined above) defining the grain boundary is constant except at the positions of the defining dislocations. The standard deviation of $h(i,t)$ falls as fluctuations of bubble size appear between the dislocations, roughening the belt of six-coordinated bubbles in these regions. The subsequent growth is broadly in line with the increasing width seen in other systems. For clarity we only show data for one type of grain boundary. For the low angle case the general form of the variation was as shown for the incommensurate grain boundary, the

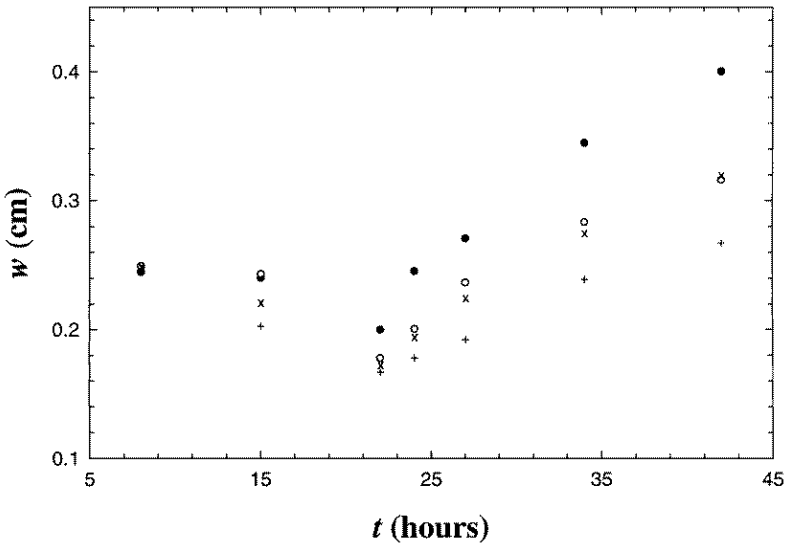

Figure 10. The variation of the roughness $w(L, t)$ for an incommensurate grain boundary. Data are shown for various samples of length L ($L = 60$, full length, ●; $L = 32$, ○; 16, ×; 8, +).

absolute values being somewhat lower, as expected.

For a self-affine interface $w(L, t)$ exhibits various scaling behaviours with both L and t [18]. We have considered these matters, averaging $w(L, t)$ for lengths $L = 32$, 16 and 8 over all possible correlated intervals $[x, x + L]$. However, while $w(L, t)$ does vary with both L and t (figure 10), in a manner reminiscent of certain self-affine systems exhibiting scaling [19], the present ranges of both parameters are too limited to enable us to ascertain scaling behaviour with any confidence. We therefore do not dwell upon these details.

4. Conclusions

We have investigated the evolution, both topological and spatial, of grain boundaries in monolayer foams. The results of both aspects are more comprehensive than the experimental data previously available [6, 13]. In all cases the 2D foam about the grain boundary evolves towards increasing disorder, as expected. For one specific system—the incommensurate grain boundary—the results are generally very similar to those from an earlier study of 2D foam containing several spatially separated dislocations [11]. Perhaps surprisingly, the evolution of those foam properties studied here did not seem to be strongly affected by differences in the angular mis-match and in bubble size between the two domains of ordered foam adjoining the grain boundary. We believe that the maxima in μ_2 observed in figure 4 are intrinsic to the evolution, rather than an artefact as for point defects [11]; this may connect with the transient peak in μ_2 found in the coarsening of relatively ordered 2D foam [6].

The major surprise of the topological part of this study is the quantitatively different behaviours observed for foams in the two cells used. Certain comparisons with previous studies [12, 13] support these differences. However, as has been remarked, they also serve to emphasize the difficulty of finding model systems which properly reproduce all features of the behaviour of a real physical system. In the present case, as noted above, the absolute values for μ_2 observed for soft boundary conditions may be more appropriate to those found

in 'normal' 2D foam [6]. This observation may suggest that studies of the evolution of point defects [10, 11, 13] might more appropriately use such a cell.

The propagation of the disorder into the ordered domains of foam adjoining the grain boundaries, and the roughening of the front between the two regions, is much as expected. While scaling of the roughening might be expected, we cannot test this hypothesis as the data do not extend to long enough times and the size of the sample is too small. While this aspect of the data is probably irrelevant to the physics of foam, it could provide an interesting example of a roughening system.

Acknowledgments

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