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Submitted on 1 Jan 1994

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Dislocation-mediated period adaptation in magnetic « bubble » arrays

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(Received 14 June 1993, accepted in final form 21 October 1993)

Abstract. — Dislocation-mediated mechanisms of stress relaxation, generating disorder in magnetic bubble arrays in response to temperature-induced period adaptation, are described. In contrast to the response of assemblies of rigid particles, the stress-induced evolution of dense magnetic domain patterns is characterized by the interplay of lattice topology and bubble domain size and shape. Such coupling of topological and geometrical degrees of freedom determines the local transformations which mediate lattice expansion (« coarsening ») and govern dislocation dynamics: these are shown to be predicated upon adjustments in the number density of bubbles via bubble collapse. Elementary processes underlying the translation of dislocations, the dissociation (« splitting ») of dislocation cores and the formation of virtual pairs as well as interstitials in the form of dislocation clusters of zero net Burgers vector are examined. Later stages of coarsening are shown to permit pattern « healing » via annihilation of interstitials. Examination of the close connection between the elementary processes of dislocation dynamics and the topological transformations of polygonal networks (« froths ») reveals the elimination of 5-sided cells to play a central role.

1. Introduction.

Magnetic domain patterns composed of stripes and bubbles may be viewed as manifestations of modulated phases which are stabilized by competing interactions [1, 2]. This simple concept has been advanced to account for domain formation in a wide variety of solids and fluids in both two and three dimensions [3, 4], and thus lends support to the view that the appearance of stable domain patterns in magnetic garnet films may be regarded as the direct analog of pattern formation in non-magnetic systems such as monomolecular amphiphilic films confined to an air-water interface where the phenomenon has in recent years generated considerable interest [5]. Within this framework, a new, macroscopic length scale, the modulation period, \( d \), is set by the ratio of pertinent interactions and may be tuned by altering that ratio via its dependence [6] on applied magnetic field, \( H \), and temperature, \( T \). The evolution of uni-directionally modulated stripe patterns from well-defined initial states into disordered (« labyrinthine ») final states has been recently examined in quantitative detail, employing digital line pattern analysis to derive robust structural attributes of labyrinths [6, 7].
Topological defects were shown to play a fundamental role in the evolution of disorder, and related considerations apply to the «topological hysteresis» of stripe patterns subjected to magnetic field-induced cycling [8] and to the defect-mediated melting of bubble lattices [9].

While not central to these instances of disordering and melting of patterns, a fundamental additional aspect of domain pattern evolution resides in the interplay of collective degrees of freedom and the associated topological defects, and the degrees of freedom associated with shape transformations of individual domains, including simple dilatation. Perhaps the most direct manifestation of this interplay is the morphological transformation between bubbles and stripes required for the transition between the corresponding modulated phases [3, 10]. The inaccessibility of the triagonal bubble phase throughout much of the \((H, T)\)-phase diagram [1, 11] may well have its origin in this coupling: only in a limited range of temperatures near the mean-field critical temperature \(T_C\) is the spontaneous nucleation of the expected bubble phase experimentally observed [11], while outside of this range the demagnetization from the saturated state leads to the appearance not of a bubble lattice, but of isolated, elongating stripes [6, 7]. The spontaneous elongation («strip-out») of an isolated bubble, generated by heterogeneous nucleation, but, at the requisite size possibly unstable to this very shape transformation [2, 12], would account for this rather drastic deviation from the mean-field phase behavior. A recent investigation of the disordering of bubble arrays and evolution of networks («froths») of polygonal bubbles during magnetic field-induced coarsening has revealed a special role of topological transformations involving pentagonal cells [13]. A further instance of the interplay between topological and geometrical degrees of freedom, involving only the adjustment in area (but not in shape) of circular bubbles, is the inherent coupling between the proliferation of dislocations and the appearance of polydispersity in triagonal bubble patterns [14]. As in the case of quenched size disorder, realized in binary mixtures of colloids [15] and other rigid particles [16], the translational symmetry characterizing the ordered solid phase is destroyed, particles of mismatched size acting as traps for dislocations [17].

The latter observations [14] were facilitated by subjecting an initially ordered, triagonal bubble pattern to «parametric coarsening» in near-zero magnetic field, a condition ensuring constant overall magnetization and thus a constant area fraction occupied by bubbles [18]. Period adjustment in response to cooling requires lattice expansion [6]. Qualitatively speaking, as \(T\) decreases below \(T_C\), the domain wall energy increases more rapidly (with an exponent related to that of the bulk correlation length) than does the demagnetizing energy (with the square of the order parameter exponent) so that the relative predominance of the former impedes the formation of interface. The consequent reduction in the density of domain walls implies an increase in the modulation period, \(d = d(H, T)\); that is, the temperature dependence of \(d\) in zero magnetic field is such that \(d_H(T)\) decreases with decreasing temperature [6]. The accumulating stress may be accommodated by the collapse of individual bubbles [19], or, as described, by the formation and motion of dislocations [14]. The focus of the present article is on the mechanisms underlying the motion of dislocations and the creation of interstitials and virtual pairs in the form of dislocation clusters of zero net Burgers vector. It is shown that the collapse of five-fold coordinated bubbles plays a fundamental role, thus introducing an element into the dislocation dynamics which is not manifest in assemblies of rigid particles, but may well be relevant in dense-packed assemblies of deformable bodies such as flexible latex spheres [20], or concentrated emulsions [21].

2. Materials and experimental procedures.

The observations reported here were made via polarization microscopy on thin, transparent films of a magnetic garnet of uniaxial magnetic anisotropy, and of composition
(YGdTm)₃(FeGa)₆O₁₂, grown on single crystal substrates of gadolinium gallium garnet (GGG) of (111) orientation to a thickness of about 13 μm. The experimental arrangement is described elsewhere [6]. An ordered initial state was produced by cooling the sample from the paramagnetic state in a small \( H \approx 1 \text{ Oe} \), constant magnetic field [6, 8, 14]. The ordered bubble lattice so produced was slowly cooled, over a period of approximately two hours, from approximately 190 °C to room temperature, 24 °C, to generate increasingly disordered patterns.

Video images were recorded on tape or disk [6, 7], to permit analysis of frame sequences in single steps (of 1/30 s), required to resolve details of bubble shape distortions and collapse mediating the motion of dislocations. Values for the modulation period, \( d \), were obtained from the position of the first harmonic in azimuthally averaged Fourier spectra computed from each pattern. Following high pass filtering and simple global binarization based on identifying the threshold with the mean of the gray scale histogram, bubble area histograms were constructed on the basis of a simple region filling algorithm which facilitates counting of interior pixels and simultaneously generates a point pattern composed of bubble centroids. Topological defects were located in the Voronoi diagram derived from this point pattern [14, 22].


Cooling from the paramagnetic state in a constant, small field produces bubble patterns such as those in figure 1, typically composed of large ordered regions (Fig. 1A) separated by grain boundaries. As with conventional polycrystalline materials, these grain boundaries are formed by chains (or arrays) of dislocations [23], as is apparent in the Voronoi diagram (Fig. 1B), and they may thus act as sources and sinks of dislocations.

![Fig. 1. — Grain boundaries observed at \( T = 184.5 \text{ °C} \approx 0.99 T_C \) in ordered initial bubble domain pattern (A), generated by cooling from the paramagnetic state in a small magnetic field [6], consist of chains of dislocations. The Voronoi diagram (B) contains 5-sided and 7-sided polygons, shaded light gray and dark gray, respectively, which mark the positions of 5-fold and of 7-fold coordinated bubbles in the original pattern. The period is approximately 9.5 μm, the horizontal dimension of the field of view in each panel is approximately 195 μm.](image)

Stress may be induced in an initially ordered domain pattern by tuning the modulation period, \( d_H(T) \), via its temperature dependence along trajectories of constant overall magnetiza-
tion in the \((H, T)\)-phase diagram \([1, 6]\). Figure 2 contains a plot of the experimentally determined temperature dependence of \(d\) for the material under consideration here. The analysis underlying the fits of the data in zero field in figure 2A is based on a scaling argument \([24]\) yielding a prediction of the form \(d_{H=0}(t) \sim t^{1/4}\), where \(t = T_C - T\) denotes a reduced temperature. To parametrize the experimental data, fits to the suggested power law with functional form \(d_H(T) = A(T_C - T)^z\) were carried out; \(A\) and \(T_C\) were varied in all fits, while the exponent, \(z\), was varied as an additional parameter, or held fixed at \(z = 1/4\) in the fits actually shown in figure 2. Fits with fixed \(z\) were based on a subdivision of the probed temperature interval, as indicated in the figure, leading to results far superior to those obtained from fits of the entire data set with floating or with fixed \(z\). Of most immediate interest to the present exposition is the obvious fact that \(d_H(T)\) exhibits the expected increase with decreasing temperature. In addition, while no claim is made as to the uniqueness of the power law representation, or to the values for \(T_C\) extracted from it, distinct regimes of temperature.

Fig. 2. — Temperature dependence of modulation period, \(d_H(T)\), determined for stripe pattern at \(H = 0\) Oe (A) and for bubble pattern at \(H = 1\) Oe (B) on the basis of azimuthally averaged 2d Fourier spectra computed from the patterns. The temperature dependence shown here is completely reversible, while the accompanying pattern morphology is generally not \([3, 6]\). As discussed in the text, solid lines in (A) were derived from fits to the functional form \(d_H(T) = A(T_C - T)^z\), with \(z\) fixed at 1/4 and respective optimal values for \(T_C\) of 209 °C and 192 °C for the two separately analyzed temperature intervals indicated in the figure. The insert to (A) also indicates that, as \(T \to T_C\), \(d_H(T)\) approaches a finite limiting value \(d_C > 0\).
dependence of $d_H(T)$ in the vicinity of the (mean-field) critical point are clearly identified. The origin of these regimes is an issue which will be revisited in the Discussion below. The lower temperature regime, probed by the data for bubble patterns in figure 2B, exhibits obvious deviations from the scaling behavior just discussed; this is not surprising in view of the garnet's ferrimagnetic nature. Pertinent to the present discussion is the continued, now evidently super-linear increase of $d_H(T)$ with decreasing temperature.

Dislocations are formed in response to the accumulating expansive strain to facilitate the requisite reduction in the number density of domains. In the case of expanding stripe patterns, the ejection of stripes by dislocation climb [6] actually suffices to maintain, albeit via disordered transients, the ordered lamellar stripe configuration. For the case of expanding bubble patterns, subjected to the condition of maintaining constant area fractions of the two coexisting magnetization states, the ordered initial state is not preserved under the imposed lattice expansion. While possible in principle via simultaneous ejection of excess rows in the three symmetry directions, this would require a directional correlation between dislocations existing in the pattern, and this is not observed, dislocations instead appearing with random orientation (modulo $\pi/3$). The ensuing evolution of disorder via proliferation of dislocations and an inherently arising polydispersity were the subject of recent quantitative analysis [14].

The condition of constant «coverage» implies simple proportionality between $\langle A_6 \rangle$, $A_6$ denoting the area of bubbles decorating 6-fold sites, and the hexagonal unit cell area of the ordered bubble pattern, $A_{\text{hex}}$, so that $\langle A_6 \rangle \sim A_{\text{hex}} \sim q^{-2}$, where $q = |q_H(T)|$ represents the modulation wave number. Figure 3 demonstrates that the mean domain area, $\langle A \rangle$, derived from an area histogram [14], also scales as $\langle A \rangle \sim q^{-2}$ This implies $\langle A \rangle \sim \langle A_6 \rangle$, an

![Graph](image)

Fig. 3. — Modulation wave number ($q$-) dependence of various geometric quantities describing magnetic bubble domain patterns. Solid lines in the plots of $\langle A \rangle^{-1}$ and of $\Delta N \equiv N - N_{\text{opt}}^{\text{opt}}$ vs. $q^2$ connect the respective rightmost point, corresponding to the ordered initial state, with the origin. Solid and dashed lines in the plot of $(\xi_0 q)^{-2}$ vs. $q^2$ serve to guide the eye. Values for $q$ were determined from azimuthally averaged 2d Fourier spectra.
observation consistent with that of constant coverage [18]. Explicit inspection actually reveals that, to better than ~ 5 %, \( \langle A_6 \rangle = \langle A \rangle \), a condition which appears surprising in view of the asymmetry of the domain area histogram [14], manifest in the difference between relative magnitudes of successive mean areas, \( \langle A_6 \rangle - \langle A_3 \rangle > \langle A_7 \rangle - \langle A_8 \rangle \), as well as in the systematic increase, \( \sigma_5 \leq \sigma_6 \leq \sigma_7 \), of the corresponding standard deviations. This point will be further considered in the Discussion.

Dislocations, generated in response to the temperature-induced strain, move to the edges or to nearby grain boundaries to facilitate the requisite reduction in the number density of domains. As the plot in the lower portion of figure 3 reveals, the disordered pattern of (fixed) linear dimension \( L_d \) maintains a roughly constant, positive excess, \( \Delta N = N(T) - N^{\text{opt}}_s(T) > 0 \), in the number, \( N(T) \), of domains relative to the « optimal » number, \( N^{\text{opt}}_s(T) \sim L_d^2/A_s \), for the corresponding perfectly ordered pattern, composed of six-fold coordinated domains of area \( A_s \). As will be described below, this excess is in fact accommodated via the formation of interstitials. The roughly constant \( \Delta N \) suggests that the pattern remains at the yield threshold, forming dislocations and eliminating (rows of) particles at a sufficient rate to track the increase in modulation period.

The average (lateral) distance between dislocations, \( \xi_D \sim n_D^{-1/2} \), determined simply by their density, \( n_D \), sets the scale for the decay of translational correlations [16, 17]. The dependence of the related dimensionless quantity \( (\xi_D/q) \sim n_D/q^2 \) on \( q^2 \) suggests the emergence of asymptotic level corresponding to \( \xi_D \approx 4d \).

The motion of an isolated dislocation through an ordered pattern, driven by expansive strain (and the resulting Peach-Kohler force), is depicted by the snapshots in figure 4. It is apparent

![Snapshots](image)

Fig. 4. — Snapshots in (A) and (B), separated in time by 1/6 s, depict the translational motion of an isolated dislocation through an ordered bubble pattern; (A) captures a transient state. The period is approximately 10 µm. A unit translational step of a dislocation is recorded in (C) and (D), separated in time by 1/15 s. The motion involves climb of one of the partial dislocations marked by excess rows terminating in the dislocation core. For each panel, the area of interest is marked by a box of which a two-fold magnification is also displayed; the black dot marking the center of each 7-fold coordinated bubble serves to guide the eye. The horizontal dimension of the field of view for each panel is 570 µm.
that the dislocation moves via climb of one of its constituent partial dislocations [23] in the direction of one of the excess rows terminating in its core. Careful inspection of the elementary translation, shown in figure 5, reveals this step to involve the elimination of the 5-fold coordinated bubble of the original dislocation core. Employing unprimed and primed numerals to refer, respectively, to domains of the indicated coordination before and after the unit

Fig. 5. — Schematic representation in (A) serves to illustrate the process underlying climb of a partial dislocation, resulting in a displacement along the vector \( \mathbf{u} \). Indicated in the original configuration are: the core (striped and hashed circles) of the dislocation of Burgers vector \( \mathbf{b} \), and the hashed cell surrounding a 6-fold coordinated bubble which will turn into a 7-fold coordinated bubble. The requisite elementary topological transformation for this translational dislocation motion, applied to the Voronoi diagram associated with the bubble pattern, involves elimination of one 5-sided cell, as sketched in (B). This topological process is detailed in the inset: solid lines represent polygonal cell edges: of these, the one marked "X" will be eliminated, leading to appearance of the two new edges indicated by dashed lines. Numerals indicate the change in the number of edges of adjacent cells [29].
translation, and defining the symbols «↓», «▲» and «▼» to signify, respectively, the operations «collapses», «contracts into» and «expands into», the process may be schematically summarized as follows:

5↓; 7▲6'; 6▼7'; 6▲5'.

In regular crystals, dislocation climb generates either vacancies or interstitials, and the requisite mass transport by diffusion generally represents the rate-limiting step [25]. The spontaneous elimination of bubbles from magnetic patterns by collapse removes the constraint of conserved interstitial transport, thereby facilitating dislocation climb. Pure dislocation glide has in fact not been observed under the experimental conditions pertinent here.

Dislocations first appear in any finite field of observation containing an ordered pattern of typically 0.5 × 0.5 μm² area via translational motion (Fig. 5) originating at sources, possibly grain boundaries, external to the observed patch. The subsequent dissociation of dislocation cores is a frequent occurrence in the early stages of coarsening. As figure 6 demonstrates, the net Burgers vector is conserved by this splitting process whose elementary steps are shown in figure 7 to involve the consumption of three bubbles.

![Dislocations](image)

Fig. 6. — Dissociation (« splitting ») of dislocation core (A) yields two dislocations (B) whose Burgers vectors sum to that of the original defect. The period of the pattern, recorded at 132 °C (= 0.87 Tc), is approximately 10 μm; the horizontal dimension of each field of view in (A) and (B) is 190 μm. The area marked in the original pattern (A)/(B) is displayed twofold magnified in the upper/lower portion of the central panel; the black dot marking the center of each 7-fold coordinated bubble serves to guide the eye.

As dislocations proliferate, clusters of dislocations begin to appear. Prevalent among these are pairs of dislocations of zero net Burgers vector, illustrated by the pattern and its associated Voronoi-diagram in figure 8. These pairs are identified in figure 9 as virtual pairs and interstitials [9, 26]. This assignment may be verified by comparing the number of bubbles
Fig. 7. — Schematic representation in (A) illustrates the process underlying the dissociation (« splitting ») of a dislocation core. Indicated in the original configuration are: the dislocation core (striped and hashed circles), the two pairs of 6-fold coordinated bubbles from each of which will emerge one 7-fold coordinated bubble, and, finally, the hashed cells surrounding 6-fold bubbles which will emerge as 5-fold coordinated bubbles. The requisite elementary topological transformation for this core splitting process, applied to the Voronoi diagram associated with the bubble pattern, may be represented in form of the (successive) elimination of three 5-sided cells, as sketched in (B): step (I) → (II) represents a translation, identical to that in figure 4; the new dislocation is generated in step (II) → (III); the final configuration is reached by another translation, (III) → (IV), of the newly created dislocation.

enclosed by a circuit about the defect pair with the number of bubbles enclosed by the same circuit in the ordered reference state: this procedure yields a vanishing differential for the virtual pair and an excess of one bubble for the interstitial; for a vacancy, not observed in the patterns discussed here, a deficit of one bubble would result.
Fig. 8. — Disordered bubble pattern and associated Voronoi diagram [14], revealing a high density of dislocation pairs of zero net Burgers vector in the form of virtual pairs (→ VP) and interstitials (→ I). Several instances of incipient unbinding of disclinations (→ d) are also apparent. The period of the pattern, recorded at 36 °C (∼ 0.66 $T_C$), is approximately 22 μm; the horizontal dimension of the field of view is 850 μm.

Virtual pairs were observed to nucleate spontaneously in magnetic bubble domain patterns subjected to shear, their ionization generating pairs of counterpropagating dislocations [27]. In the present experiments, virtual pairs formed as the result of the recombination of dislocations of opposite Burgers vector, a process facilitating the elimination of excess (half) rows. The core of the resulting virtual pairs represents a metastable configuration. As indicated in figure 9, its elimination requires a redistribution of edges between adjacent 5-sided and 7-sided polygons in the associated Voronoi diagram. This edge exchange is identical to the (edge and hence cell conserving) T1 elementary transformation mediating the coarsening of polygonal networks, or froths [13, 28]. Virtual pairs were found to persist inspite of being unstable to this process. The requirement for bubble size adjustment, a geometrical response to be made concurrently with the topological edge exchange, may be cited as a likely impediment to the recombination of virtual pairs.

Interstitials accommodate the excess of domains, $\Delta N$, contained in disordered patterns relative to the ordered reference state [14]. The annihilation of these defect configurations is therefore predicated upon the removal of domains via bubble collapse. As illustrated in figure 10, the annihilation of interstitials by just such a collapse process, accompanied by the (non-conserved) transfer of magnetization from the vanishing 5-fold to an emerging 6-fold bubble, is actually observed in the « later » stages of coarsening. Interstitial annihilation is readily accomplished by the elimination of a 5-sided cell from the Voronoi polygon associated with the bubble pattern. This process, detailed in figure 11, is identical to that postulated [29] for the elimination of 5-sided cells from magnetic froths of polygonal bubbles subjected to increasing applied field where it appears to control the rate of coarsening [13]. Given the
Virtual Pair
(\(\delta N = 0\))

Interstitial
(\(\delta N = +1\))

Fig. 9. — Schematic representation of virtual pair (A) and interstitial (B), each formed by a pair of dislocations of zero net Burgers vector. Open, striped and hashed circles respectively represent 6-fold, 7-fold and 5-fold coordinated domains. Also indicated (« → ») are CCW circuits about the defect pair employed to establish the mismatch, \(\delta N\), in the number of enclosed domains relative to that expected for the ordered reference state. Sketched below each defect pair is the topological transformation which, when applied to the Voronoi polygon associated with each bubble pattern, would eliminate that defect. Required for the annihilation of the virtual pair in (A) is the T1 process of edge exchange, while the simplest local transformation to remove the interstitial in (B) is the elimination of one 5-sided cell [29] (see also Fig. 10).

temporal resolution (of 1/30 s) afforded by standard video recording, the collapse of one 5-fold bubble is virtually indistinguishable from the «symmetrical» alternative, namely the coalescence of two 5-fold coordinated bubbles in the interstitial. Applied to the associated Voronoi diagram, coalescence corresponds to the inverse of mitosis, or polygon («cell») division [28].

The elimination of topological defects, e.g. via the annihilation of interstitials described here, would facilitate a return to an ordered state of the pattern. A necessary condition for the occurrence of this particular process appears to be the requirement that the area, \(A_6\), of the one 6-fold bubble replacing the two 5-fold bubbles not exceed the maximal area consistent with 6-fold coordination. This condition apparently prevents the annihilation of interstitials at high number densities of bubbles in the initial stages of disordering.

4. Discussion.

The formation of ordered patterns of given modulation period, \(d = d(H, T)\), in a sample of linear dimension, \(L_0\), requires the realization of the correct number density, \(n \sim L_0/d\), of stripes or \(n \sim (L_0/d)^2\) of bubbles whose correct size, i.e. width in the case of infinite stripes, and area,
Fig. 10. — Snapshots, separated in time by 1/30 s, recording the elimination of an interstitial from a disordered bubble pattern; the middle panel captures a transient state. The area marked in the original pattern is displayed twofold magnified in the right hand portion of each panel. The period of the pattern, recorded at 36 °C ($= 0.66 T_c$), is approximately 19 μm; the horizontal dimension of the left hand portion, containing the original pattern, is 285 μm.

Annihilation of Interstitial ($\Delta N = -1, \Delta e = -1$)

Fig. 11. — Schematic representation illustrating the topological transformation mediating the annihilation of an interstitial. The process relies on the elimination of a 5-sided cell from the Voronoi diagram associated with the bubble pattern (see Fig. 4), resulting in a net change in the number of domains of $\Delta N = -1$, and in the number of polygon edges of $\Delta e = -1$.

$A_o$, in the case of circular bubbles, is determined by the requisite value of the overall magnetization. Consequently, the evolution of ordered triagonal bubble patterns under the global constraint of constant net magnetization implies $A_o \sim d^2$, because each bubble occupies a constant fraction of the hexagonal unit cell [14]. Under these conditions, period adjustment introduces stress in dense-packed stripe and bubble arrays, and this may be accommodated via formation and motion of defects. A simple process of stripe rupture and dislocation climb...
mediates relaxation in lamellar stripe domain patterns, facilitating preservation of the state of lamellar ordering via formation of transiently disordered stripe patterns [6]. In contrast, bubble patterns generally evolve into disordered states of hexatic order via the proliferation of dislocations [14].

The pattern evolution of interest here occurs in response to adjustments in the modulation period, \( d = d_H(T) \), in a manner following the dependence described in connection with figure 2. Fits to the functional form \( d_H(t = T_C - T) \sim t^{1/4} \), shown in that figure, confirm the expected increase of \( d_H \) with decreasing temperature over most of the accessible range pertinent to the pattern evolution described here. However, in the vicinity of the (mean-field) critical point, striking deviations from this behavior manifest themselves.

First, it is apparent that \( d_H(T) \to d_C = 0 \) as \( T \to T_C \). This behavior was in fact predicted [24] on the basis of considering the interaction between increasingly diffuse domain walls, also expected in the regime of weak segregation near the consolute point of binary mixtures. While a quantitative treatment remains to be given, the existence of a finite limiting value for the modulation period as \( T \to T_C \) suffices to lead one to suspect a peak in the susceptibility \( \chi_q \) of the disordered phase, i.e. above \( T_C \), at the finite \( q = 2 \pi / d_C \), as observed in low temperature dipolar Ising ferromagnets [30]. The existence of such a peak for a planar system of azimuthal symmetry would imply profound fluctuation-induced modifications of the mean-field critical behavior, including the elimination of the critical point, and the suppression of the critical temperature [31].

Such a lowering of \( T_C \) could in fact account for the existence of distinct regimes of temperature dependence of \( d_H(T) \) apparent in figure 2A. This conclusion is suggested by the fits, displayed in that figure, of \( d_H(t) \) to a power law with a fixed exponent of \( z = 1/4 \): this analysis would indicate that the primary distinction between the two regimes of figure 2 is a sudden drop (of approximately 15°C according to the fits) in the effective transition temperature, \( T_C \). There is, however, an alternative explanation, invoking a transition between Bloch and Ising domain wall structures [24, 32]. Laser Bragg scattering experiments would offer a possible experimental approach to ascertain the effects of fluctuations above \( T_C \) and to help resolve the origin of the observed behavior.

The evolution dynamics of magnetic bubble patterns is characterized by a close interplay between topology and geometry. The topology of magnetic bubble patterns is faithfully represented by the Delaunay triangulation, a planar graph containing only vertices of degree three centered on the bubble centroid positions, and representing the dual of the Voronoi diagram, associated with a given bubble pattern [22, 28]. The set of bubble areas represents a scalar field defined on the triangulation and must therefore respect the constraints so imposed. Magnetic bubble patterns arrange in such a way that each domain adjusts so as to occupy all locally available space while maintaining a finite, minimal separation between domain boundaries [33], the latter condition representing a manifestation of repulsive dipolar interactions between domains [1, 2]. An immediate implication of this observation [33] is the fact that the availability of dilatation, i.e. area adjustment, and eventually of shape distortions [3], as additional, geometrical degrees of freedom avoids the open areas associated with « jammed » configurations characterizing random packings of rigid disks [34].

The area of a given bubble cannot be adjusted without eventually adjusting bubble coordination. Thus, for given net magnetization, or bubble « coverage », bubble coordination, \( n \), and bubble area, \( A \), are not independent variables, but are instead subject to conditions governing static pattern configurations as well as evolution dynamics. Polygonal froths display an interdependence of shape and size of polygonal cells described by a joint probability distribution, \( p(n, A) \) of cell coordination and area [28, 35]. A corresponding distribution should apply to the Voronoi diagram associated with a bubble pattern, with the consequence
that larger bubbles display higher coordination. In support of this view we have the trimodal bubble area histogram characterizing disordered patterns [14], restricting the form of the requisite distribution \( p(n, A) \) so that \( \langle A_6 \rangle - \langle A_5 \rangle > \langle A_7 \rangle - \langle A_6 \rangle \) for the mean areas, \( \langle A_6 \rangle = \int A p(n, A) \, dA \), of \( n \)-fold coordinated bubbles, and \( \sigma_5 \approx \sigma_6 \approx \sigma_7 \) for the corresponding variances. In addition, the patterns of interest here conform to the conditions \( N_5 = N_7 \) and \( N_5 + N_7 + N_6 = N \) expressing the fact that topological defects appear, over the range of disorder investigated here, exclusively in the form of pairs of (±1) disclinations, respectively marked by 5-fold and 7-fold coordinated bubbles. Invoking these conditions, it is readily shown [36] that \( \langle A \rangle - \langle A_6 \rangle = - c_D(2 \langle A_6 \rangle - \langle A_5 \rangle + \langle A_7 \rangle) \). That is, the difference between \( \langle A_6 \rangle \) and \( \langle A \rangle \) is determined by the number density of defects, \( c_D = N_5/N = N_7/N \) and by the curvature of the function \( \langle A_6 \rangle = \langle A_6 \rangle(n) \). The sign of the asymmetry in the mean areas cited above implies that the latter is in fact finite and positive for the distribution \( p(n, A) \) pertinent to the patterns investigated here. With typical numbers extracted from the most disordered pattern encountered here [14], i.e. \( c_D \approx 0.25 \), \( \langle A_5 \rangle / \langle A_6 \rangle \approx 0.593 \) and \( \langle A_6 \rangle / \langle A_7 \rangle \approx 0.774 \), one indeed finds that \( \langle A \rangle - \langle A_6 \rangle < 0 \). The explicit form of an appropriate \( p(n, A) \) for a disordered bubble pattern would have to take into account the expected \( n \)-dependence of the ratio \( \langle A_6 \rangle / \langle A_6^{\text{Vor}} \rangle \) of the mean area of an \( n \)-fold bubble and that of the \( n \)-sided Voronoi polygon, although such a \( p(n, A) \) only specifies near-neighbor correlations and will likely not be unique [36].

A topological characteristic of the evolution dynamics of magnetic bubble patterns investigated here is the conservation of the triangulation. More specifically, only topological excitations of unit charge (±1) appear in the temperature range accessed by the pattern evolution reported here. These topological restrictions fundamentally limit the set of possible pattern configurations, eliminating in particular configurations otherwise observed in planar patterns such as the droplet patterns produced by vapor deposition on surfaces [37] and notably the disordered bubble patterns frequently encountered in Langmuir films [4, 38] whose topology cannot be represented by a planar graph [33]. Within the framework of a recently proposed model for the evolution dynamics of dense bubble patterns, the existence of a topological conservation law and the absence of spontaneous, temporal coarsening, a property associated with magnetic bubble patterns, are intimately related [33].

The evolution of disorder in dense-packed bubble patterns is mediated by the proliferation of dislocations, coupled with the polydispersity arising from the availability of bubble dilatation as an additional degree of freedom to accommodate stress [14]. The disordered pattern may be viewed as a « frustrate » state of the system for given field, \( H \), and temperature, \( T \) : while an ordered ground state realizing the correct modulation period, \( d_H(T) \), is always well-defined, relaxation to this « reference » state is prevented by an excess of bubbles, \( \Delta N \), and by the presence of topological defects generated in the course of period adaptation. As with the evolution of labyrinthine stripe domain patterns from an initial lamellar state [6, 7], bubble pattern evolution would seem to be primarily determined by the requirement to realize the correct modulation period for given \( H \) and \( T \). That is, the emerging pattern morphology may be regarded to represent the constrained optimization of the pertinent free energy functional [1, 2, 6, 7] under the condition of fixed coverage and given number density of bubbles, and hence in the presence of a preset concentration of topological defects.

The intriguing possibility of enabling processes which contribute to a « healing » of a given disordered pattern by deleting topological defects and thereby facilitating a relaxation into the ordered reference state is in marked contrast to the coarsening of polygonal froths from an initially ordered state : in that situation, the existence of a single topological defect in the form of a 5-7 pair of polygons may trigger the destabilization of the entire pattern [13, 28, 35, 36]. It
will be interesting to see whether suitable constraints may be placed on model evolution
dynamics [33, 35] to investigate the possibility of re-entrance into ordered states. On the
experimental side, examination of more advanced states of disorder than those realized in the
present study will have to reveal whether such reentrance does in fact occur, or whether
additional mechanisms of topological disorder, for example in the form of disclination
unbinding (Fig. 8), predominate the further evolution of disorder into an amorphous state [14,
16, 17].

Acknowledgements.

The work presented here has benefited from conversations with R. Seshadri, C. Volkert and
R. Wolfe, from a variety of incisive observations offered by M. Magnasco, and from the
comments of a pair of critical reviewers.

References

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and 5150.
[18] With the exception of the symmetry axis $H = 0$ of the $(H, T)$ phase diagram, trajectories with
constant $H$ do not preserve overall magnetization. However, the trajectory $H = 1$ Oe, followed
in the present experiments, was found, via evaluation of histograms, to closely preserve the
area fraction of minority («white») bubble phase at a value of 45% ± 2%, indicating the
overall magnetization to remain approximately constant as well.
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