

Polymorphic phase transitions in systems evolving in a two-dimensional discrete space

A. Gadomski

*Department of Theoretical Physics, Institute of Mathematics and Physics, University of Technology and Agriculture,
85-796 Bydgoszcz, Aleja Kaliskiego 7, Poland*

(Received 3 November 1998; revised manuscript received 22 February 1999)

Polymorphic phase transitions in systems evolving in a two-dimensional discrete space have been studied. The driving force of the transitions appears to be a difference between two main energetic contributions: one, related to the thermal activation of the process, and another, being of quantum nature. The former (high temperature limit) is naturally assigned to the expansion (melting) part of the transition, while the latter (low temperature limit) has much in common with the contraction (solidification) part. Between the two main physical states distinguished, there exists a certain state, corresponding to a discontinuity point (pole) in the morphological phase diagram, represented by the well-known Bose-Einstein (Planck) formula, in which the system blows up. This point is related to an expected situation in which the contour of the object under investigation stands for the Brownian or purely diffusional path, with the fractal dimension $d_w=2$, and the situation can be interpreted as some emergence of an intermediate “tetratic” phase. This, in turn, recalls a certain analogy to the equilibrium (order-disorder) phase transition of Kosterlitz-Thouless type, characteristic of, e.g., rough vs rigid interfaces in a two-dimensional space, with some disappearance of interface correlation length at $d_w=2$. Otherwise, the contours of the objects are equivalent to fractional Brownian paths either in superlinear or “turbulent” ($d_w<2$; the expansion case), or sublinear, viz., anomalously slow ($d_w>2$; the contraction case) regimes, respectively. It is hoped that the description offered will serve to reflect properly the main subtleties of the dynamics of the polymorphic transitions in complex “soft-matter” systems, like formation of lipid mesomorphs or diffusional patterns, with nonzero line tension effect. [S1063-651X(99)08706-1]

PACS number(s): 05.70.Fh, 71.10.-w, 05.60.Cd, 81.30.Fb

I. INTRODUCTION

A main challenge of recent developments is the recognition of the interplay of microscopic interfacial dynamics with external macroscopic fields in the determination of the morphology of evolving patterns. The real challenge, however, appears when a system under study is complex, i.e., it possesses quite nontrivial dynamics [1]. This is very often the case of many biophysical systems, like, e.g., lipid membranes, liquid-crystalline assemblies, protein crystals, model biomaterials, etc., which manifest high viscosity, sensitivity to some changes of external physicochemical conditions (temperature, pressure, pH, light, strain-stress response, etc.), chemical reactivity, presence of fluctuations of either thermal or athermal nature; moreover, since we may also consider a behavior of individuals of nanometer sizes and in very short time (low temperature) scales, a certain role of quantum effects cannot be excluded *a priori* [2].

The scaling concept (more generally, the renormalization-group approach) [3], being a fundamental theoretical idea of a microscopic-to-macroscopic adjustment of systems composed of a certain number of elementary subunits (molecules; individuals), was frequently used to characterize the behavior of such systems in both static (time-independent) as well as dynamic domains. Quite often the random walk concept was attached to it [3], being in the last few decades intensively developed in the both above mentioned directions; see [1,4]. The concepts listed above have often been accompanied by the concept of anomalous kinetics in complex systems, usually named the glassy dynamics or dispersive (in some sense, fractal) kinetics [5].

The phase changes in “soft-matter” systems, e.g., col-

loids, surfactants, complex fluids, amphiphiles, etc., having their prerequisites in some rather old but perennially alive critical phenomena, like ferromagnetic-to-paramagnetic phase transitions or transformations in superfluids (^4He), are presently a subject of intense study (cf., [6,2], and references therein). An intriguing task here seems often to be a reasonable though phenomenological (even) description of the system under study in terms of some heterogeneous (invoking this notion, we wish to stress an important role of the interface or quasicrystal’s surface, or finally, of the grain boundaries) or, in particular, polymorphic phase transition concept, being applicable to a “soft-matter” system, where a thermotropic, barotropic, as well as lyotropic mesomorphism (for example, of phospholipids) appears to be a key feature thereof [7].

While trying to describe the phase changes we have to know whether they take place near the equilibrium point or rather out of it (sometimes also: how far from it). We have to know as well whether we will be interested in some static picture of the process, such as, for example, diagrams of coexisting phases, or perhaps we wish to know, how might the phases intermingle if, for instance, the temperature is changed, and how does the process look just in the course of time? It is also a possibility to work in a certain “combined” way. E.g., we can benefit from some knowledge of the (static) equilibrium phase picture, just for attempting to use it effectively in constructing the so-called morphological (dynamic) phase diagrams. This method will be exemplified by certain known scenarios, in which some emergence of (dis)ordered shapes under nonequilibrium growth conditions, like various mesomorphs in lipids [7], Hele-Shaw or “diffusion-limited-like” patterns in the so-called dense-

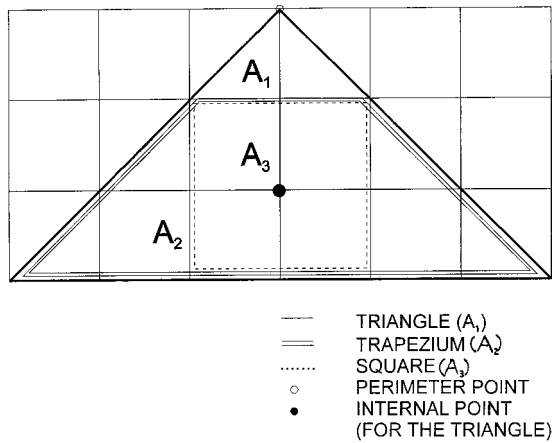


FIG. 1. Schematic representation of the Steinhaus rule (2). The areas of the triangle, trapezium, and square are designated by A_1 , A_2 , and A_3 , respectively, and are evaluated to be 9, 8, and 4; the internal as well as perimeter points of the triangle have been drawn as well (for the other symbols, see the legend of the picture).

branching morphology regime, etc. [8–13]. Moreover, to get a physically consistent picture we do postulate a kind of thermodynamic-geometrical adjustment of the system under study, just to let the complex system evolve at criticality in a self-organized manner [14].

In this work, to study both the nondynamic as well as dynamic aspects of polymorphic phase transitions in a two-dimensional (2D) discrete space, we propose a (simple) model that merges a discrete picture of a polygon embedded in the square lattice with some thermodynamic-geometrical concept. The thermodynamics means here certain scaling arguments [12], while geometry is “reduced” to use a Steinhaus rule for evaluating the polygon’s area (Fig. 1) [15]. Since the main goal of this paper is mostly to reveal a general mechanism leading to the determination of the “averaged” velocity of the evolving ensemble (cluster of molecules; “molecular condensate;” quasicrystal) while passing (slowly or vigorously) through many states of quasiequilibria, e.g., in ordered spin glasses [5], the dynamics of the process will be of interest. Yet, some equilibrium properties of the contraction-expansion (specifically, solidification vs melting) phase transformation will be mentioned, too. Especially, we wish to recall the role of structural defects [6] when considering the transition process (see Appendix A).

Thus, as is probably expected [5,13], by imposing a discrete time dynamics on the system under investigation and working within the concept of existence of a set of dynamic quasiequilibria (and also by assuming that the system is capable of passing through them), we are able to get some evolution rules for the surface fractal in a 2D space (square lattice). The surface (enveloping line) is proposed to be modeled by a random walk trajectory, which is a fractal of dimension d_w [4]. The process is, in general, highly curvature driven, which is very characteristic of many phenomena of biological interest, like solidification (crystallization) in complex media, wetting, formation of biomembranes (vesicles, micelles), bubbles, and interfaces [7,10,12].

The paper is organized as follows. In the next section, we present a picture of the growing process in a discrete 2D space (the reader is also encouraged to see [13]). In Sec. III,

some basic steps of construction of the morphological phase diagram are proposed, whereas in Sec. IV its basic properties in terms of the synchronized dynamics (collective or cooperative dynamics) have been revealed, and the physical origin, touching somehow the quantum character of the thermally activated transition process, has been pointed out. In this section, the thermodynamic-geometrical adjustment mentioned above has been utilized (it provokes discussion of the self-organization or some other type of effectiveness of the growing process [14]). As a result, a Bose-Einstein-like relation has been derived (see also Appendix B) and discussed in terms of the so-called Kosterlitz-Thouless (KT) order-disorder phase changes (cf. Appendix A), for which both the dynamic structural adjustment and discreteness of the physical space play an important role [16]. Conclusions regarding some examples based preferentially on studying the lipid, and similar, e.g., bacteria systems [6,7,11], complete the investigations presented in this work.

II. DISCRETE PICTURE OF THE PROCESS

Certain physically valuable information is accumulated about a cluster (molecular “condensate” or crystal), at a dynamic quasiequilibrium with its surroundings and located in a 2D space (square lattice) on its nucleus, if one provides (i) an “averaged” statistical-thermodynamical description in terms of the scaling concept, like

$$\langle A \rangle \sim p^{D_{ws}}, \quad D_{ws} \in [0, \infty), \quad D_{ws} \neq 1, \quad p \gg 1 \quad (1)$$

where p stands for the number of the perimeter (or periphery) sites of the polygon of averaged area $\langle A \rangle$ embedded in a 2D space; the periphery cluster’s sites are the most outer occupied sites of the object under consideration, and D_{ws} represents the random walk size exponent [cf. [9,12,13], for some examples; the scaling formula (1) has proved to be useful at least for, e.g., vesicles or Eden clusters, with $D_{ws} \approx 3/2$, i.e., approximated by the *self-avoiding random walk* (SARW) trajectory, or diffusion-limited aggregates (DLA) grown under vanishing surface tension conditions, with $D_{ws} \approx 1$, but not precisely equal to 1 [10,11,8]]; (ii) exact information about the area A (not to be confused with $\langle A \rangle$) of a convex single polygon (cluster) embedded in the square lattice, and spanned on a certain number of lattice nodes that are either the internal [denoted by i ; here $i \geq 1$, cf. Eq. (1)] or the external (perimeter) points, designated by p (again), found by Steinhaus [15] to be (see Fig. 1; for convincing the reader, some examples with a triangle, trapezium, and square have been offered)

$$A = i + \frac{p}{2} - 1. \quad (2)$$

To enter the physics of the cluster (polygon) at the equilibrium point, let us, for example, suppose that both p and i sites, placed on the square lattice nodes, are occupied exclusively by lipid molecules, and that the centers of inertia of the macromolecules correspond to the precise locations of p ’s and i ’s. Note also that the molecules undergo, independently and with their own frequencies, oscillations around the position of the centers of inertia at equilibrium. Invoking a simplest and well-known approximation, they can be rec-

ognized as harmonic oscillators (Appendix B), though some extension to the anharmonic case is plausible. (Note that the oscillating ensemble as a whole does oscillate with an average frequency, being the arithmetic mean of individual eigenfrequencies.) Going back to Eqs. (1) and (2), let us accept in our further studies some negligibly small statistical uncertainty in the system (“compact” lattice objects grown from a nucleus; no fuzzy or “dispersive” aggregates), namely, $\epsilon \equiv |\langle A \rangle - A|$, for a certain ϵ so that $0 < \epsilon \ll 1$, which results [by comparing directly Eqs. (1) and (2)] in

$$i \equiv i(p) = ap^{D_{ws}} - \frac{p}{2} + 1, \quad (3)$$

with $D_{ws} \in [0, \infty)$, but $D_{ws} \neq 1$, e.g., $D_{ws} \approx 3/2$ [12]. Note that a is a positive proportionality (thermodynamical) parameter. It can be called the area-expansion (or, contraction) parameter [see Eq. (1), in which a may always enter as a prefactor]. The bigger a is the larger the averaged area of the cluster can be. If, in turn, a attained smaller values, then the cluster would shrink by decreasing its area. Let us notice that a must depend upon temperature and may vanish at a critical temperature for which $D_{ws} = 1$ so that Eq. (3) will be useless if taken for further analysis because i 's can be negative, even though p will be the smallest, i.e., when $p = 4$ [look at Eq. (2): if $p = 4$ and $i = 0$, then $A = 1$, and the possibly smallest cluster in the square lattice consists exclusively of molecules located at its periphery].

Relation (3) stands for some equilibrium global characteristics of the cluster formation process the mechanism of which has not yet been specified. It is taken at a single dynamic (*sic*) quasiequilibrium point. It goes without saying, however, that the usefulness of relation (3) in such a form can easily be questioned, e.g., because of its incompleteness, which means that we do not know the locations of i and p points, so that we are (at this stage of our considerations) not able to reconstruct how the cluster looks, just from the information contained in Eq. (3). But, we believe that the “statistical information measure” given by Eq. (3) is properly constructed, at least, because it is straightforwardly obtainable from Eqs. (1) and (2), and because it includes the desired thermodynamic-geometrical content that we wish to have. Perhaps, an appealing simplicity is here the main argument for keeping Eq. (3) in our further study; see [13], for further analysis of Eq. (3).

Now, let us make the two next physically motivated assumptions. The first is that the system under study approaches not one single dynamic quasiequilibrium state, but rather passes, in subsequent time instants t_1, t_2, \dots , through, in general, infinitely many states of dynamic equilibria so that it is eventually driven out of the single equilibrium from which we start at $t_0 \geq 0$. We are here mostly motivated by well-known facts concerning either extremally vigorous or apparently slow transport as well as relaxation dynamics in many “soft-matter” systems, like polymers, model biomaterials, (ordered) glassy or polycrystalline systems, etc. [3,5,4]. The second is, in fact, a presumption that the scaling form (1) simply survives for a quite broad range of growing processes [1,11,12], no matter how far the physicochemical conditions controlling the process, e.g., temperature, will be changed.

In consequence, all the above stated points result in the following: (i) i and p must be functions of time, i.e., $i \equiv i(t_n)$ and $p \equiv p(t_n)$, for $n = 0, 1, 2, \dots$; (ii) if the system leaves one dynamic equilibrium state (at t_{n-1} , for example) for another neighboring state (at t_n), then the prefactor a as well as the (dynamic) exponent D_{ws} may change their values because the move is certainly caused by a change of the physicochemical conditions controlling the cluster formation process; in this case, we have to write down explicitly that $a \equiv a(p_{\text{term}})$ and $D_{ws} \equiv D_{ws}(p_{\text{term}})$, where p_{term} is generally related to a control parameter taken from the parametric set of physicochemical conditions of the cluster formation process. Let us notice, in this moment, that by making use of assumptions (i) and (ii) in our further studies, we are going to postulate something like the existence of a certain equivalence between temperature (thermodynamic parameter) and time (temperature-time or Williams-Laudel-Ferry equivalence principle [5,1]) since any sufficiently detectable change in value of the control (thermodynamic) parameter should cause a “jump” of the system towards another dynamic equilibrium state detected at time t_n , where $n = 1, 2, \dots$. Such a situation is very characteristic of, e.g., polymeric systems in polycrystalline or amorphous states of characteristic relaxation (Vogel-Fulcher-Tammann) time [3,5,2]; see Appendix B for further discussion.

III. MAIN STEPS OF CONSTRUCTION OF THE MORPHOLOGICAL PHASE DIAGRAM

If a general picture drawn in Sec. II is accepted, let us from now try to push the system (molecular agglomerate) through the dynamic quasiequilibria, or to impose a discrete time dynamics on it. In other words, we will be interested in knowing what is the total number of the internal subunits of the cluster at time instant t_n [denoted by $i(t_n)$], having known that at the preceding time moment t_{n-1} their number is equal to $i(t_{n-1})$. We can obtain it, if we simply perform discrete differentiation over both sides of Eq. (3), which results in

$$\frac{\Delta i}{\Delta t} = \left(a D_{ws} p^{D_{ws}-1} - \frac{1}{2} \right) \frac{\Delta p}{\Delta t}, \quad (4)$$

where $\Delta i = i(t_n) - i(t_{n-1})$ and $\Delta p = p(t_n) - p(t_{n-1})$; obviously, $\Delta t = t_n - t_{n-1}$. Some discussion of the derived equation in both discrete (Δt finite and sufficiently small) as well as continuous ($\Delta t \rightarrow 0$) regimes has been presented in [13] for a case mostly pertinent to the formation of vesicles, where the surface (in a 2D space, one has a line “enveloping” the object under growth) was modeled by a SARW trajectory. Notice that for a close-to-equilibrium case, in turn, one gets $\delta p / \delta i = (a D_{ws} p^{D_{ws}-1} - \frac{1}{2})^{-1}$, where δp and δi stand for very small departures of p and i from the corresponding equilibrium values, and one may expect a certain number of physically interesting effects, like existence of some “tetratic” (intermediate) phase, provided that there exist two main types of phases (a low temperature contracted, for $D_{ws} < 1$ and a high temperature expanded, when $D_{ws} > 1$), and disappearance of the coherence length, or even certain indications of a topological defects’ mediated transition [16] (see Appendix A for details).

Now, let us be interested in a more general case far exceeding the case for which $D_{ws} = D_{SARW}$, where $D_{SARW} \approx 4/3$ [9]. Namely, we wish to consider a quite general physical situation

$$D_{ws} = \frac{2}{d_w}, \quad (5)$$

where d_w represents a dimension of the random Brownian path that is either of pure Brownian nature, with $d_w = 2$ (i.e., our singularity point on the nonequilibrium morphological phase diagram that we are going to invent), or the surfaces of the objects are equivalent to fractional Brownian paths in superlinear, viz., extremely fast (say, turbulent; $d_w < 2$), or sublinear, i.e., anomalously slow ($d_w > 2$) regimes, respectively; see [4,5] for some basic information.

Throughout the rest of the paper, we wish to explore, according to what has been proposed in [8] and also by others [10,11], some analogy between equilibrium phase diagrams, like that presented in, e.g., [12,6], and the nonequilibrium morphology diagrams offered for systems with the shapes which are observed to change their characteristics, from being more or less ordered (quasicircular) to exposing a rather irregular (disorder) peripheral microstructure (DLA-like fractal, seaweed, or dendritic objects (see [10]) in the case when the driving force of the process is slowing down, e.g., Hele-Shaw patterns, or systems manifesting the so-called dense-branching morphology); see Ben-Jacob and Garik in [8] for details. In particular, in their studies, the authors [8] propose to examine relationships between an ‘‘averaged’’ growth velocity (designated by α), i.e., the velocity ‘‘weighted’’ according to the geometric-dynamical characteristics of the interface [in our model, the relative changes in p stand for the excess quantity, when compared with the changes of i of the same type; see Eqs. (6) and (7) below], and the control parameter, just representing some quantitative characteristics of the surface of the evolving object [in our case, it must be either D_{ws} or d_w ; see Eq. (5)]. The velocity α will be derived in a way similar to that proposed in [8], i.e., $\alpha = \alpha_p / \alpha_i$, where the two α ’s are defined as

$$\alpha_p = \frac{p^{-1} \Delta p}{\Delta t}, \quad \alpha_i = \frac{i^{-1} \Delta i}{\Delta t}. \quad (6)$$

Rearranging Eq. (4) and using $\alpha = \alpha_p / \alpha_i$ as well as recalling Eq. (6), we obtain

$$\alpha \equiv \alpha(D_{ws}) = \frac{2i}{p} \frac{1}{2aD_{ws}p^{D_{ws}-1} - 1}. \quad (7)$$

This way, we get a certain prerequisite of the morphology (dynamic) phase diagram $\alpha(D_{ws})$ which can be drawn for respective time instants t_0, t_1, t_2, \dots . The first thing that must be noticed, however, is that looking at Eq. (7), one sees a possibility or even danger of some morphological catastrophe (escape of α to ∞). This can surely happen if [see Eq. (7), again]

$$2aD_{ws}p^{D_{ws}-1} = 1. \quad (8)$$

It is equivalent to extract a quantity, designated by p_{mc} , which one may call a critical number of perimeter points attained if the system would undergo a first order phase transition (or, specifically, a kind of piezoelectric effect could be noticed, if one presumed that we are working with ferroelectric 2D quasicrystals), with some release of a transition heat, and when the system eventually arrives at one of the two main transition states discontinuously (a sharp peak in α), and if there is no chance to detect any intermediate state (see discussion above). In other words, one could state that the first order phase transition cannot be ruled out. This can certainly happen at a temperature (transition) point T_{mc} . Thus, the above mentioned quantity looks like

$$p_{mc} = (2aD_{ws})^{-1/D_{ws}-1}, \quad a > 0, \quad D_{ws} \in [0, \infty), \quad D_{ws} \neq 1. \quad (9)$$

Let us state explicitly that the first morphological smash ($\alpha \rightarrow \infty$) assigned to the evolving system is to carry it towards a global (contrary to infinitely many states of dynamic quasiequilibria which can be anticipated as local) equilibrium point, assigned either to a solidified (collapsed or shrunken) or to a molten (expanded or extended) state. We may name this type of catastrophe shock undercooling or shock overheating, respectively, depending on the direction of the driving force, e.g., undercooling [10,11]. Following the rationale expressed above, however, we should like to keep p_{mc} constant because global equilibrium characteristics have to be more or less balanced by a play of parameters driving the system. There is also such a possibility in our description. It can be realized by noticing that if one observes some change in D_{ws} , caused by a change in the physicochemical conditions of the process, then it must be an adequate (natural) compensation of this effect by changing a , and vice versa, so that for $D_{ws} \neq 1$, one would assume that there exists a limiting constant value of p_T ,

$$p_T \equiv (2aD_{ws})^{-1/D_{ws}-1} = \text{const} > 0. \quad (10)$$

Let us mention that in this way we have, in some sense, worked out the thermodynamic quantity p_T to be independent of an interplay between D_{ws} and a , i.e., of the dynamic interface characteristics when compared with the expansion or contraction magnitude of the cluster. Note, however, that by doing that some constraints have been imposed on the system; cf. Appendix A. Keeping this in mind, one can rewrite Eq. (7) as

$$\alpha(D_{ws}) = \frac{2i}{p} \frac{1}{q_T D_{ws}^{-1} - 1}, \quad (11)$$

where $q_T = p/p_T$ stands for a kinetic-thermodynamical quotient. There exists another factor being rather of kinetic-geometrical nature. Let us denote it by $q_G \equiv 2i/p$ [see Eq. (11)].

Now, we wish to introduce a next (topological) simplification of our description. It relies on considering exclusively the geometrical objects spanned on the nodes of the lattice that are of quasicircular form (see discussion in [13]). In consequence, we take into account such 2D objects which are quasicircles of radius $r \pm \mu(\phi) \delta \pm \nu(\phi) \delta^2 \pm \dots$, where δ , the perturbation amplitude, being usually very small when

compared with r , and μ , ν are continuous perturbing functions of angular argument ϕ ; cf. [17,18].

Next, let us simplify the system even more. This means, let us assume that the evolving quasicircular object is of constant density. Under such an assumption, the constant number density of internal points of the cluster, denoted by ρ_i , is given by $i/\pi r^2$ while the constant number density of perimeter points of the cluster, ρ_p , can be written as $p/2\pi r$. Using these trivial arguments, one can write explicitly

$$q_G \equiv \frac{2i}{p} = \frac{p}{p_G}, \quad (12)$$

where

$$p_G = \frac{2\pi\rho_p^2}{\rho_i}, \quad (13)$$

and is constant. (Notice that, for the sake of clarity and simplicity, we have assumed that the evolving object is quasicircular, as e.g., in the case of Mullins-Sekerka or Saffman-Taylor instability concepts [11] which have proved to be useful, at least, in the description of the crystal or fingered growth.) Using Eq. (13) we can rewrite our basic relation as

$$\alpha(D_{ws}) = \frac{q_G}{q_T^{D_{ws}-1} - 1}. \quad (14)$$

Note that q_G , like q_T , is also independent of D_{ws} and a , and that both q 's are dynamic (they implicitly depend upon time) as well as dimensionless variables.

At the end of this section, let us realize that another morphological catastrophe may appear, when looking at formula (14), namely,

$$D_{ws} = 1, \quad (15)$$

which corresponds to a situation in which the interface ‘‘molecular condensate’’ vs surroundings is represented by the pure Brownian path (a most disordered or ‘‘disorganized’’ case). In this case, we have again to observe some escape of α to ∞ . This type of morphological crash we will rather assign to the continuous phase transition concept, with no release of the transition heat, but with a certain appearance of strong microstructural disorder at the interface. This kind of disorder may lead to a detection of some intermediate phase which can be called the ‘‘tetratic’’ phase (Appendix A), by a certain verbal analogy with the well-known hexatic phase, mostly observed in melting phenomena realized in a triangular lattice. Notice that formula (14) is the most general relation representing the morphological phase diagrams that we offer in our present description, and that the jump of α to infinity may appear for either the first order ($D_{ws} \neq 1$, but with appearance of p_T) or the continuous ($D_{ws} = 1$, exclusively) phase changes. In particular, it may throw more light on the classification of the transitions in all the systems undergoing diffusional growth, with nonvanishing surface (line) tension effect [10,8,17,18]. It would also be of use when studying the polymorphic phase transformations in phospholipids [7,6,1], especially when some terminology borrowed from physical metallurgy, like the martensitic or diffuse phase transitions, may enter [7].

IV. POSTULATE OF SYNCHRONIZED EVOLUTION DYNAMICS: EMERGENCE OF A BOSE-EINSTEIN ‘‘MOLECULAR CONDENSATE’’

In this section, we propose another very interesting and physically motivated simplification to be built into our approach. Namely, we postulate that [cf., Eqs. (10) and (13)] p_T and p_G can take on a common value p_c , namely,

$$p_c \approx p_T = p_G, \quad (16)$$

which results in $q \approx q_T = q_G$ (let us believe that such a q exists). This can be called the assumption (or postulate) of the synchronized dynamics, which means that the kinetic description proposed has reached a kind of thermodynamic-geometrical *consensus*. It may happen when the process in question proceeds gradually rather (like in chemical reactions, for example, where the system quite frequently [14] gets self-organized) than under some ‘‘hard’’ physical conditions. So, we claim here, in fact, that the process goes in a self-organized (i.e., possibly efficient) way. We see that it may be the easiest way for the system to evolve. It reflects also our opinion that the complex physical systems behave quite ‘‘economically’’ when they follow their evolution under the sometimes very cumbersome influence of physico-chemical factors causing certain changes in the system behavior, i.e., at the vicinity of a critical parameter point p_T ; cf. Eqs. (16) and (10). In other words, by postulating Eq. (16), we assume that the system near criticality organizes itself to pass smoothly through the ‘‘landscape’’ of dynamic quasiequilibria, which is very characteristic of, e.g., biological processes [19b]. Another rough but quite general interpretation may be proposed: the system probably ‘‘sees’’ that it is (energetically) better for it to proceed in an evolutionary rather than a revolutionary way.

Assuming Eq. (16) (with all the consequences of doing that) one gets immediately

$$\alpha(D_{ws}) = \frac{q}{q^{D_{ws}-1} - 1}. \quad (17)$$

Let us notice that we have perhaps surprisingly gotten, e.g., a formula that resembles very closely the Bose-Einstein (BE) distribution of phonons in an insulator [20]. This is even its generalization. Obviously, some applications of the Bose-Einstein or Planck formula much exceed the case mentioned. It is more general. E.g., it concerns the superconductivity (mostly of second type) as well. Generally speaking, it describes the behavior of some superfluid or superconducting condensates. Do we have also in our case behavior which resembles, at least in part, a BE condensation (e.g., an ensemble of atoms oscillating around their equilibrium positions, and eventually spreading out or collapsing, as is expected in the case studied)?

Before really trying to answer this question let us proceed further in that field. Namely, utilizing Eq. (16), let us make use of the (discrete) time-temperature equivalence principle mentioned in Sec. II. In other words, let us ‘‘measure’’ q in terms of discrete time instants or in terms of temperature values assigned to the subsequent states of dynamic quasiequilibria. It can be done, e.g., for a finite number of measurements, and when one states in a naive way that

$$q = e^{\pm 1/n}, \quad n = 1, 2, \dots, N \quad (18)$$

where $e \approx 2.71$ stands for the Napier constant and N is a natural number that can be interpreted as a last time or temperature quantity measured (do not forget that $q = p/p_c$). The \pm sign preceding the exponent n^{-1} is introduced here to distinguish between the two main growing (quite general) situations that we immediately recognize: the system either collapses (shrinks, solidifies, or “coagulates”) in the course of time [“−” sign in the argument in Eq. (18); $D_{ws} < 1$] or expands [gets molten or swollen; “+” sign in the argument in Eq. (18); $D_{ws} > 1$]. [Note that in the contraction regime, and when the time-temperature equivalence principle is fulfilled [5], Eq. (18) stands for a Boltzmann energy term.] There is also a certain case “in between,” i.e., when the evolution of the system is stopped or disturbed somehow, e.g., by extremal freezing or heating, or interfacial structural breakdown ($D_{ws} = 1$) assigned to the interfacial or surface behavior of the evolving object. These situations correspond to a blow up of α mentioned above. Notice here that by assuming Eq. (18) the number of perimeter sites p is measured either as a multiple of p_c , which corresponds to some expansion of the agglomerate, or as a fraction of p_c , which indicates the contraction or shrinkage. Combining Eqs. (17) and (18), and multiplying both sides of the resulting equation by $e^{\mp 1/n}$, one can accurately arrive at the BE formula (cf. [20], Chap. 6 or [19])

$$\alpha'(D_{ws}; n) = \frac{1}{e^{\pm(D_{ws}-1)/n} - 1}, \quad n = 1, 2, \dots, N \quad (19)$$

where, instead of α , an exponentially weighted quantity $\alpha' = \alpha e^{\mp n}$ (the prime means that this quantity is primed, not differentiated) was introduced. For a more convenient analysis let us rewrite Eq. (19) in a compact as well as more readable form, namely,

$$\alpha'(z) = \frac{1}{e^{1/z} - 1}, \quad (20)$$

where $z^{-1} = \pm \Delta E_s / E_T$, and $\Delta E_s = D_{ws} - 1$ as well as $E_T = n$ (n 's indicate the subsequent time instants of dynamic equilibria taken at corresponding temperature values). The first energetic term, ΔE_s , corresponds to a change of the interfacial free energy (related very much to the surface tension) which is either released to the surroundings (with “+” sign; the expansion case) or taken from it (with “−” sign; the contraction case). Because the time-temperature equivalence principle is fulfilled, the second energetic term, E_T , corresponds to the Boltzmann thermal energy.

A few remarks seem to be appropriate here. First, note that $|z| > 1$, which means that for a small free energy change, $\alpha' = z$ (for a given n), i.e., a linear case appears, which is even the case of $z \sim 1$ [20]. If, in turn, $|z| \ll 1$, we get a nonlinear (of large free energy change) $\alpha' = e^{-1/z}$, also for a given n .

Second, as was mentioned before, the expansion dynamics is taken with a plus sign ($+\Delta E_s$) and because of $D_{ws} > 1$ [4], in this case, z remains positive. If, in turn, the contraction (shrinkage) dynamics is realized, the change of interfacial free energy is of opposite sign, but fortunately,

$D_{ws} < 1$, so that the argument z also takes on positive values [20]. (It also means that there is, in fact, no reason to take absolute values of z .)

Thus, we have arrived at a more or less consistent picture of the phase transition during the evolution process that we propose to study in terms of the morphological phase diagram $\alpha'(z)$. It represents a relation between an “averaged” [8] and “thermally weighted” [see Eqs. (19) and (20)] velocity α' of the evolution of a molecular cluster in a 2D discrete space and the interfacial free energy excess (inevitably associated with a change of surface tension [19]), represented by z , realized during the phase transformation.

The morphological phase diagram shows that there are two evolving phases: an expanding (specifically, molten) ($\Delta E_s > 0$ and $D_{ws} > 1$ or $d_w < 2$) and a collapsing (say, solidified or “frozen”) ($\Delta E_s < 0$ and $D_{ws} < 1$ or $d_w > 2$); cf. Eq. (5). The important case “in between” corresponds to a “morphological crash” at the interface [see Eq. (15)], which mathematically means that $\alpha'(z)$ blows up (the main mathematical reason is that $D_{ws} = 1$ or $d_w = 2$, which is equivalent to the fact that the interface growing object–external medium is a pure Brownian trajectory). This scenario enables us also to recall some rationale about the presence of the intermediate phase (“tetratic” phase) [21]. Thus, the existence of a discontinuity (pole) at $D_{ws} = 1$ in $\alpha'(z)$ makes a choice for interpreting it in terms of KT order-disorder phase change; see Appendix A for further discussion. If one defines a characteristic interface length or a correlation length, say θ as $\theta = z^{-1}$ (but taken also per unit interface length), then one may notice that θ vanishes, just at $d_w = 2$ at some transition temperature point. This again resembles quite apparently the scenario usually assigned to KT phase transitions [16] (best known for 2D Coulomb lattice gas, where the transition between an “ordered” state of “frozen dipoles” and a plasma disordered state is noticed), especially for systems such as rigid vs rough interfaces in a 2D space [22]. Such transitions are classified to be of infinite order according to the Ehrenfest scheme; see [16] for details. Moreover, let us note that the analytical description of the transformation process is given, when the synchronized dynamics is presumed, by the BE relation (see Fig. 6.2 in Chap. 6 of [20]), much more pronounced in the expansion than in the contraction case (for the latter, $D_{sw} \in [0, 1)$, exclusively), or its generalization; cf. Eqs. (14) and (16). The synchronized dynamics would be a landmark of self-organized criticality (see, the beginning of the section) and would manifest some defense of the system against undergoing the phase transition of the first order. Under the assumption of relatively “lazy” dynamical behavior, observed, e.g., in (ordered) spin glasses as well as in biosystems (i.e., under some presence of constraints and hierarchy of degrees of freedom, from slow to fast) [5, 1, 2, 21], one can arrive at the Vogel-Fulcher-Tammann characteristics, and its relations to the quantum ground state averaged energy of the ensemble as a whole; see Appendix B.

V. CONCLUSIONS

In this study, a phenomenological approach to the polymorphic phase transition in complex systems [23], where the structure formation process takes place on a seed located somewhere in a square lattice, has been proposed. The main

idea was to apply both the scaling argumentation as well as a planimetric Steinhaus rule, within some level of uncertainty, and to postulate that the system passes, in principle, through infinitely many dynamic quasiequilibrium points, like in the “long-tail” or dispersive kinetics concept in biophysics [24,5]. The result is that a nonlinear relationship [Eq. (3); see Sec. II] has immediately been recovered, and by imposing a (discrete) time dynamics on the system, one is able to arrive at an evolution equation [difference scheme; cf. Eq. (4)] that includes inherently a curvature change term [13] very characteristic of the growing phenomena, like crystallization (also, single crystalline domains in lipid monolayers [23]), solidification (faced 2D quasicrystals [2,18]), emergence of bubbles and vesicles and microdomain growth [17,2,21], formation of biomembranes (interfaces) [9,25], etc. It is worth stating here explicitly that the description offered is based on utilizing the random walk concept, which has proved to be useful in this subject [12,9,4,3].

The procedure for the morphological phase diagram invented in Sec. III apparently shows that the kinetics of the growing process can only very crudely be interpreted (see Binney *et al.* [16], Chap. 1) as that of first order [see Eq. (10); the discontinuity at p_T as well as some common expectation that the transition heat is released suggest doing so], like that of diffusional growth; cf. [10], and references therein. But we see a more subtle and self-consistent interpretation of the transition process that we have studied. Namely, it resembles very much order-disorder phase transitions of KT type (see above), e.g., those for rigid (ordered) vs rough (disordered) interfaces in two-dimensional (discrete) space [22]. The characteristic (or correlation) interface length θ , inversely proportional to z (see Sec. IV), vanishes right at the point of disorder picked up at $D_{ws} = 1$, or equivalently, $d_w = 2$.

To be more specific, and for convincing the reader of the usefulness of our approach, a few examples have been juxtaposed below. These are (i) as reported by Ben-Jacob and co-workers [8], mostly for Hele-Shaw patterns [18], and by Brener, Müller-Krumbhaar, and co-workers [10], there exist very complex dense-branching morphologies (DBM) that, under nonequilibrium growth conditions, produce objects with ordered shapes, and the accompanying interfacial pattern formation process includes certain selection rules; the same was confirmed in [10], where some morphology diagrams for diffusional growth [17] have been sketched, and where, as a result of interplay between the driving force (undercooling) and the surface anisotropy, a variety of morphologies, from, e.g., compact seaweed, via some intermediate fractal dendritic to compact dendritic have been obtained (yet, the processes have been preferentially assigned to the first order phase transition processes, though a chaotic regime has also been mentioned) [10]; (ii) comparing to (i), similar patterns have been obtained, and analogous scenarios have been drawn by Möhwald and co-workers [23] for amphiphilic monolayers (with plenty of structural, e.g., fractal forms therein), Ben-Jacob and co-workers [8], and Matsushita and co-workers (they also deliberated about some application of the random walk concept) for bacteria, also Nittmann and co-workers [11] for 2D myristic acid monolayers, where a passage with characteristic surface development, commencing from a liquid condensed phase, “skip-

ping” over an intermediate phase to eventually ending at the liquid expanded state, along with increase of temperature, has been reported; (iii) as impressively illustrated by Laggner, Kriechbaum, and co-workers [7], the thermotropic or barotropic phospholipid phase transitions may result in a multitude of polymorphic forms, such as, e.g., lamellae or hexagonal patterns, and there are some processes that conform to the scenario sketched in the paper, like the lamellar-to-inverse hexagonal (H_{II}) transition or even a pretransition in phosphatidylcholines, where, in the course of temperature change, a positional order of macromolecules is gradually lost (an orientational order is at least slightly perturbed), and where an intermediate order-disorder zone (with its disclination lines) emerges during the process; and (iv) Gruner [25] and others [6] also pointed out some interesting approaches and ideas, mostly related to the concept of the curvature (in the spirit of Helfrich [6]) of model biomembranes; there, one considers the normal micellar-bilayer-inverse micellar interpassage as a good candidate conforming to the expansion (head groups outwards) vs contraction (head groups inwards) phase transformation, with the “tetratic” intermediate (head groups in a plane) phase in between.

Listing the above examples, we may firmly say that there is here a place for BE statistics as a tool for describing the evolution process in terms of the morphological diagram that we propose. It is so, indeed, since the BE statistics is the quantum statistics, and specifically, the velocity α' [compare Eqs. (14), (20), and (B4) in Appendix B] emerges as a consequence of a difference of the two energetic contributions, i.e., one related to the thermal excitations of the cluster’s molecules, and another one, which is the averaged energy of the quantum ground states; cf. Appendix B. In other words, the fundamental “engine” of the process is a difference between activities of the thermal phonons of the “molecular condensate,” associated somehow with oscillations of individual molecules, and the dislocational phonons “stuck” to the groups of molecules, constituting structural defects [21,26,27] of the condensate; cf. Eqs. (B4), (B5), (A5), and (A6) in Appendixes A and B, for comparison. (By the way, note that some quite strict relations between the density of states of phonons in solid crystals, and the anomalous random walk concept, given by a walk visiting distinct sites of the crystal, which is a key feature of the dispersive kinetics, see Płonka in [5], have been presented, e.g., by Bunde in [18].) Moreover, a selection mechanism naturally operates during the transition process (see Appendixes A and B), since the dislocations (*viz.*, disclination pairs, characterized by their Frank index [21,26]) as well as the quantum (Planck) time scale effects do enter. Thus, by making use of this observation, and realizing the elementary driving force [Eq. (B4) in Appendix B] of the process, under the dispersive (“long-tail”) kinetic regime [24], we may attempt to elucidate various morphologies mentioned above.

Finally, let us underscore that it is not the first physical situation [3,4] in which the random walk concept may prove useful, and that it provides some reasonable argumentation for elucidating complex phenomena.

ACKNOWLEDGMENTS

A few ideas related to this study have been presented at the XI Marian Smoluchowski Symposium on Statistical

Physics, Zakopane, Poland. Support from Polish State Committee of Scientific Researches is acknowledged.

APPENDIX A: LANDMARKS OF THE CONTRACTION VS EXPANSION KOSTERLITZ-THOULESS-LIKE PHASE TRANSITION

It was noticed at the beginning of Sec. III that for a close-to-equilibrium case one provides

$$\frac{\delta p}{\delta i} = \left(a D_{ws} p^{D_{ws}-1} - \frac{1}{2} \right)^{-1}, \quad (\text{A1})$$

where δp and δi stand for very small departures of p and i from the corresponding equilibrium values. [Note that the right-hand side of Eq. (A1) depends exclusively upon one of two main dynamic variables, that means, upon p , which will also be helpful for specifying the transition as being mediated by a disclination line [26].] After applying a boundary condition [16], i.e., $\delta p/\delta i = 1$ (approximately) around $p = p_c$ to Eq. (A1), where p_c [recall Eq. (16)] will provide the renormalization (scale magnification) factor, and after rewriting the resulting equation in a logarithmic form, one gets [see Eq. (3), again]

$$(D_{ws} - 1) \ln p_c + \ln D_{ws} - \ln \frac{3}{2a} = 0. \quad (\text{A2})$$

Even a very crude analysis of Eq. (A2) may lead to a certain number of physically interesting effects, like a simple analytical demonstration of the presence of some ‘‘tetratic’’ (intermediate) phase, and disappearance of the coherence length, or even certain indications of a topological defects’ mediated transition. Since necessary details can be found elsewhere [16,21,26], it will be shown below in a sketchy way.

1. Simplified D_{ws} renormalization: Existence of a ‘‘tetratic’’ phase and disappearance of the coherence length

After making use of Taylor expansion for $\ln D_{ws}$ up to third order, i.e., $\ln D_{ws} \cong D_{ws} - 1 + \frac{1}{2}(D_{ws} - 1)^2$ since D_{ws} may also be greater than one, one physically interesting solution to Eq. (A2) has to be recast, namely,

$$D_{ws} = \sqrt{1 + \ln d_c} - \ln p_c \quad (\text{A3})$$

[$d_c = p_c(3/2a)^2$], which, after linearizing the square root in Eq. (A3) (let us call it the linear approximation, designated by D_{ws}^l ; it may be more suitable for the expansion case rather, unless some adequate compensation of d_c by p_c is provided, which is going to favor the contraction case, in turn) reads

$$D_{ws}^l = 1 + \ln \frac{d_c^{1/2}}{p_c}. \quad (\text{A4})$$

The critical point $D_{ws} = 1$ corresponds to a critical $a_c = 3/2\sqrt{p_c}$ which both mathematically (hexagonal shift) as well as physically [7] means that one can ‘‘feel’’ the presence of the ‘‘tetratic’’ (‘‘hexatic’’) phase. E.g., after Laggner *et al.* [7], if the first (Bragg) [1,0] d_B spacing were equal to

$\sqrt{p_c}$, then $1/a_c$ would be related to some characteristic distance in a hexagonal lipid phase of the model 2D biomembrane. If, in turn, one defined a coherence length of the system, κ , as $\kappa \propto 1/a$, and wished to look after a critical exponent ν of the transition process, $\nu = \ln(1/p_c)/\ln(dD_{ws}/d\kappa)$ at $\kappa_c = 1/a_c$ and for $p_c \rightarrow \infty$, one obtains at once some striking loss of coherence because $\nu = \infty$. Obviously, one must be aware of the very crudeness of the approximation offered here.

2. Topological defects’ mediated transition:

Similarity to a crystal’s disclination approach for nematics due to Landau and Lifshitz

If one cares about mechanical singularities that support KT transitions well [16,21], one can notice that Eq. (A1) is exactly of the form of that used by Landau and Lifshitz to describe an effect of homogeneous disclination for nematics [26] [see Eq. (37.2) in the seminal book of Landau and Lifshitz, and the analysis thereafter]. Transforming the results presented in [26] to our language, we should notice that here a ‘‘configurational disorder’’ of p and i points or some crowding (a loss of positional and a perturbation of orientational orders) of the internal and mostly peripheral points will be responsible for a change of the disclination (Frank) rank m . (Recall that if $m = 0$, i.e., the index vanishes, the structure of the crystal is not unperturbed mechanically.) Under the assumption that the total number of points in the crystal, $j = i + p$, changes periodically according to a rule [26], like $j(i + p_c) = j(i) + 2p_c D_{ws}$, one provides

$$m = 1 + \left(a p_c^{D_{ws}-1} - \frac{1}{2} \right)^{-1}, \quad (\text{A5})$$

which would mean that the disclinations were generated mostly at the periphery of our 2D crystal, and that they grew or shrank as a result of some competition between the expansion/contraction magnitude, given by a and the ‘‘renormalization factor,’’ represented by p_c ; cf. Sec. IV. One can also explicitly write down that

$$D_{ws} = 1 + \frac{\ln[(m+1)/2a(m-1)]}{\ln p_c}. \quad (\text{A6})$$

Note that by comparing Eq. (A6) with Eq. (A4) the linear approximation mentioned above also provides a as a function of m and p_c . For completeness, it should be stated that m can be either positive or negative, and cannot take any real value, but the multiples of $1/2$, or must be an integer [26]. In that sense, the ‘‘spectrum’’ of D_{ws} is definitely not a continuous spectrum. This way, we agree with another constraint of the transition process [5].

APPENDIX B: HARMONIC APPROXIMATION TO THE THERMAL ACTIVATION OF THE ‘‘MOLECULAR CONDENSATE’’—QUANTUM EFFECT AND VOGEL-FULCHER-TAMMANN BEHAVIOR

The class of processes that we are going to describe here is undoubtedly a type of process that is driven by a difference between some macroscopic and microscopic fields. As pointed out by Schöck [27], the contribution of the macro-

scopic field is often equivalent to thermal activation [26] [given by a thermal energy contribution $\epsilon(\omega, n)$, n stands for the temperature here] of the ensemble, which is important for the high temperature limit of the process, whereas near the low temperature limit of the cluster (“molecular condensate”) formation, one may expect the quantum effects, at a first approximation given, in the semiclassical approach, by the averaged energy of the ground states, $\epsilon_Q = (h/2\pi)(\omega/2)$ (h represents the Planck constant, and ω will be specified below), to be of prior importance [27,26,16].

Assuming that the quantum contribution may enter [27], there is, in our opinion, a place for taking into consideration the following natural redefinitions [cf. Eqs. (11) and (12)], namely,

$$q_T = e^{\pm 1/n}, \quad (\text{B1})$$

$$q_G = \frac{h}{2\pi} \omega, \quad (\text{B2})$$

where n is the temperature (time) variable frequently mentioned above (recall the time-temperature equivalence principle, cf. [5]), and ω stands for the averaged frequency of the ensemble taken as a set of harmonic oscillators; cf. Sec. III. Notice that, by introducing a dissociation energy term, some anharmonic contribution is also possible [27]. The redefinitions (B1) and (B2) enable us to present Eq. (14) (see, Sec. IV for comparison) as follows:

$$\alpha = \frac{(h/2\pi)\omega}{e^{\pm(D_{ws}-1)/n} - 1}, \quad (\text{B3})$$

which straightforwardly implies [27] that α is given by the difference between the two types of energetic contributions mentioned before, namely,

$$\alpha = \epsilon(\omega, n) - \epsilon_Q, \quad (\text{B4})$$

i.e., some elementary flux-force [14] relation is recovered. Moreover, it is easy to evaluate the walk (enveloping line) dimension D_{ws} to be

$$D_{ws} = 1 \pm \frac{h}{2\pi} \omega, \quad (\text{B5})$$

where, as before, the “+” sign corresponds to the expansion case, while the “−” sign represents the contraction case. It is worth comparing Eq. (A6) with Eq. (B5) to really anticipate the quantum contribution to the process under study. Let us bear in mind that the “tetratic” case of $D_{ws} = 1$ occurs when either $\omega = 0$ (a completely “frozen,” i.e., unrealistic case) or the “quantal” time t_Q , being of order of the Planck constant, is sufficiently small. Since the latter is very well fulfilled, one can try to conclude that the emergence of the “tetratic” phase is mostly caused by the quantum contribution, which is true in the low temperature limit (cf. [21,16], and references therein), that means, for $D_{ws} < 1$.

Moreover, it is interesting to report that for the low temperature limit, by comparing q_T and q_G [see Eqs. (B1) and (B2)], one easily recovers the well-known Vogel-Fulcher-Tammann [5,28] relaxation time τ_V for a partly (at least) disordered system, simply by noticing that

$$\omega = \omega_0 e^{-1/n}, \quad (\text{B6})$$

where $\tau_V = \omega^{-1}$, and $\omega_0 = (h/2\pi)^{-1}$. This means that this slowly varying anomalous relaxation behavior is mostly manifested in the low temperature limit (the contraction case). In particular, it would be a case of the relaxor high T_c ferroelectric films, in which this behavior is observed, being, for example, responsible for a “diffuse” phase transition, where there is a nearly continuous spectrum of transition points, as in our situation, where a multitude of dynamic quasiequilibria is present [28].

Let us also state clearly that the oscillating part of the total energy of the molecular ensemble, E_{osc} , related to the attraction potential of the disclinations [16], is quantized [see Eqs. (A6) and (B5)], and realize the contribution of Frank index m], namely,

$$E_{osc} = \pm \frac{\ln[(m+1)/2a(m-1)]}{\ln p_c^2}. \quad (\text{B7})$$

From Eq. (B7) it clearly follows that the quantization of E_{osc} is assured by m [26], whereas a [see Eqs. (1)–(3) again] accounts for whether we are near the contraction regime or rather beyond it. Note that at $D_{ws} = 1$ (“tetratic” phase), E_{osc} gets a singularity, because $a = 0$.

-
- [1] Proceedings of the First International Conference on Scaling Concepts and Complex Fluids, Copanello (Italy), 1994, edited by F. Mallamace [Nuovo Cimento D **16**, No. 7-9 (1994)]; R. Lipowsky, in *Random Fluctuations and Pattern Growth: Experiments and Models*, edited by H.E. Stanley and N. Ostrowsky (Kluwer, Dordrecht, 1988), pp. 227–245.
- [2] A. Ulman, *An Introduction to Ultrathin Organic Films. From Langmuir-Blodgett to Self-Assembly* (Academic Press, Boston, 1991), pp. 305–338; *Physics of Biomaterials: Fluctuations, Selfassembly and Evolution*, Vol. 322 of *NATO Advanced Study Institute Series E: Applied Sciences*, edited by T. Riste and D. Sherrington (Kluwer, Dordrecht, 1996).
- [3] P.-G. De Gennes, *Scaling Concepts in Polymer Physics* (Cornell University Press, Ithaca, NY, 1979); M.E. Fisher, in *Fundamental Problems in Statistical Physics*, edited by E.G.D. Cohen (North-Holland, Amsterdam, 1985).

- [4] S. Havlin and D. Ben-Avraham, *Adv. Phys.* **36**, 695 (1987); H.C. Berg, *Random Walks in Biology* (Princeton University Press, Princeton, NJ, 1983).
- [5] A. Płonka, *Annu. Rep. Prog. Chem., Sect. C: Phys. Chem.* **94**, 89 (1998); R.G. Palmer, D.L. Stein, E. Abrahams, and P.W. Anderson, *Phys. Rev. Lett.* **53**, 958 (1984); C. Dasgupta and H. Sompolinsky, *Phys. Rev. B* **27**, 4511 (1983); H. Sher, M.F. Shlesinger, and J.T. Bendler, *Phys. Today* **1**, 26 (1991); A. Gadomski, *Mod. Phys. Lett. B* **11**, 645 (1997).
- [6] J.N. Israelachvili, *Intermolecular and Surface Forces with Applications to Colloidal and Biological Systems* (Academic Press, London, 1985), Chap. 16; W. Helfrich, in *Physics of*

- Defects*, Proceedings of Les Houches Session XXXV, edited by R. Ballian, M. Kleman, and J.-P. Poirier (North-Holland, Amsterdam, 1981), pp. 716–755; M.W. Tate, E.F. Eikenberry, D.C. Turner, E. Shyamasunder, and S.M. Gruner, *Chem. Phys. Lipids* **57**, 147 (1991).
- [7] M. Kriechbaum and P. Laggnner, *Prog. Surf. Sci.* **51**, 233 (1996); P. Laggnner and M. Kriechbaum, *Chem. Phys. Lipids* **57**, 121 (1991); O. G. Mouritsen, *ibid.* **57**, 179 (1991); K. Pressl, K. Jorgensen, and P. Laggnner, *Biochim. Biophys. Acta* **1325**, 1 (1997); A. Gadomski and J. Luczka, *Acta Phys. Pol. B* **28**, 1827 (1997).
- [8] E. Ben-Jacob and P. Garik, *Physica D* **38**, 16 (1989); E. Ben-Jacob, P. Garik, and D. Grier, *Superlatt. Microstruct.* **3**, 599 (1987); E. Ben-Jacob, O. Schochet, A. Tenenbaum, I. Cohen, A. Czirok, and T. Vicsek, *Nature (London)* **368**, 47 (1994); N. Samid-Merzel, S.G. Lipson, and D.S. Tannhauser, *Phys. Rev. E* **57**, 2906 (1998).
- [9] D. Stauffer and N. Jan, *Can. J. Phys.* **66**, 187 (1988); D. Stauffer and D.P. Landau, *Phys. Rev. B* **39**, 9650 (1989); P.D. Gujrati, *ibid.* **27**, 4507 (1983).
- [10] E. Brener, H. Müller-Krumbhaar, D. Temkin, and T. Abel, *Physica A* **249**, 73 (1998); E. Brener, H. Müller-Krumbhaar, and D. Temkin, *Phys. Rev. E* **54**, 2714 (1996); E. Brener, K. Kassner, H. Müller-Krumbhaar, and D. Temkin, *Int. J. Mod. Phys. C* **3**, 825 (1992); T. Ihle and H. Müller-Krumbhaar, *J. Phys. I* **6**, 949 (1996).
- [11] K.A. Suresh, J. Nittmann, and F. Rondelez, *Europhys. Lett.* **6**, 437 (1988); M. Ohgiwari, M. Matsushita, and T. Matsuyama, *J. Phys. Soc. Jpn.* **61**, 816 (1992); A. Hill, *Nature (London)* **348**, 426 (1990).
- [12] M.E. Fisher, A.J. Guttmann, and S.G. Whittington, *J. Phys. A* **24**, 3095 (1991); C.J. Camacho and M.E. Fisher, *Phys. Rev. Lett.* **65**, 9 (1990).
- [13] A. Gadomski and Ch. Trame, *Acta Phys. Pol. B* (to be published).
- [14] I. Prigogine and G. Nicolis, *Self Organization in Nonequilibrium Systems* (Wiley, New York, 1977); W. Ebeling, *Struktur- bildung bei irreversiblen Prozessen* (Teubner Verlagsgesellschaft, Berlin, 1976) (in German); S. Boettcher and M. Paczuski, *Phys. Rev. E* **54**, 1082 (1996).
- [15] H. Steinhaus, *Kalejdoskop Matematyczny* (Wydawnictwo Szkolne i Pedagogiczne, Warsaw, 1989) (in Polish); there is also an English edition of the book: H. Steinhaus, *Mathematical Snapshots* (Oxford University Press, New York, 1979).
- [16] J.J. Binney, N.J. Dowrick, A.J. Fisher, and M.E.J. Newman, *The Theory of Critical Phenomena. An Introduction to the Renormalization Group* (Clarendon Press, Oxford, 1992), Chap. 13; J.M. Kosterlitz and D.J. Thouless, *J. Phys. C* **6**, 1181 (1973); M. Paczuski and M. Kardar, *Phys. Rev. B* **43**, 8331 (1991).
- [17] J.S. Langer, *Rev. Mod. Phys.* **52**, 1 (1980); W.W. Mullins and R.F. Sekerka, *J. Appl. Phys.* **34**, 323 (1963); J. Luczka, A. Gadomski, and Z.J. Grzywna, IMA Report 869, University of Minnesota, Minneapolis, 1991; A. Gadomski, *Vacuum* **50**, 79 (1998).
- [18] D.A. Kessler, J. Koplik, and H. Levine, *Adv. Phys.* **37**, 255 (1988); A. Bunde, in *Festkörperprobleme (Advances in Solid State Physics)*, edited by P. Grosse (Vieweg, Braunschweig, 1986), Vol. 26, p. 113; B. Berge, L. Faucheux, K. Schwab, and A. Libchaber, *Nature (London)* **350**, 322 (1991); A. Gadomski and Z.J. Grzywna, in *Proceedings of the Second International Symposium on Far-From-Equilibrium Dynamics of Chemical Systems, Świdno (Poland), 1994*, edited by J. Popielawski and J. Górecki (World Scientific, Singapore, 1991), pp. 416–427.
- [19] (a) M. Toda, R. Kubo, and N. Saito, *Statistical Physics I. Equilibrium Statistical Mechanics* (Springer-Verlag, Berlin, 1983), Chaps. 3 and 4; (b) C.C. Bradley, C.A. Sacket, J.J. Tollett, and R.G. Hulet, *Phys. Rev. Lett.* **75**, 1687 (1995); (c) H. Anderson, J.H. Ensher, M.H. Matthews, C.E. Wieman, and E.A. Cornell, *Science* **268**, 198 (1995); (d) D.R. Nelson, *Phys. Rev. Lett.* **60**, 1973 (1988).
- [20] C. Kittel, *Introduction to Solid State Physics* (Wiley and Sons, New York, 1966), Chap. 6, and references therein.
- [21] D.R. Nelson and B.I. Halperin, *Phys. Rev. B* **19**, 2457 (1979); W.F. Brinkman, D.S. Fisher, and D.E. Moncton, *Science* **217**, 693, (1982); R. Bausch, R. Schmitz, and Ł.A. Turski, *Phys. Rev. Lett.* **73**, 2382 (1994); D.E. Moncton and R. Pindak, *ibid.* **43**, 701 (1979); E.M. Chudnovsky, *Phys. Rev. B* **40**, 11 355 (1989).
- [22] V. Privman, *Int. J. Mod. Phys. C* **3**, 857 (1992); K.K. Mon, S. Wansleben, D.P. Landau, and K. Binder, *Phys. Rev. Lett.* **60**, 708 (1988).
- [23] A. Miller, W. Knoll, and H. Möhwald, *Phys. Rev. Lett.* **56**, 2633 (1986); H. Möhwald, R.M. Kenn, K. Kjaer, and J. Als-Nielsen, in *The Structure and Conformation of Amphiphilic Membranes*, edited by R. Lipowsky, D. Richter, and K. Kremer (Springer-Verlag, Berlin, 1992), pp. 9–18; K. Lohner, O.V. Konovalov, I.I. Samoilenko, I.V. Myagkov, V.I. Troitzky, and T.I. Berzina, *Thin Solid Films* **288**, 262 (1996); G.H. Peters, S. Toxvaerd, N.B. Larson, T. Bjornholm, K. Schaumburg, and K. Kjaer, *Nuovo Cimento D* **16**, 1479 (1994); G. Henn, M. Stamm, H. Poths, M. Röcker, and J.P. Rabe, *Physica B* **221**, 174 (1996).
- [24] J.F. Nagle, *Biophys. J.* **63**, 366 (1992); Yu.A. Berlin, N.I. Chekunaev, and V.I. Goldanskii, *Chem. Phys. Lett.* **197**, 81 (1992); H. Sompolinsky, *Phys. Rev. Lett.* **47**, 935 (1981); A. Gadomski, *J. Phys. II* **6**, 1537 (1996); J. Morgenstern, *Phys. Rev. B* **27**, 4522 (1983).
- [25] S.M. Gruner, *J. Phys. Chem.* **93**, 7562 (1989); *Proc. Natl. Acad. Sci. USA* **82**, 3665 (1985).
- [26] L.D. Landau and E.M. Lifshitz, *Elasticity Theory* (Nauka, Moscow, 1987) (in Russian), Chap. 6; J. Friedel, *Dislocations* (Pergamon Press, Oxford, 1964), Chap. 3; R. Holyst, *Phys. Rev. Lett.* **72**, 4097 (1994).
- [27] G. Schöck, in *Dislocations in Solids*, edited by F.R.N. Nabarro (North-Holland, Amsterdam, 1980), pp. 65–71; H. Haken and H.C. Wolf, *Molecular Physics and Elements of Quantum Chemistry* (Springer-Verlag, Berlin, 1995), Chaps. 1 and 10.
- [28] A.K. Tagantsev, *Phys. Rev. Lett.* **72**, 1100 (1994); A.A. Bokov, *Solid State Commun.* **90**, 687 (1994); Z. Surowiak, D. Czekaj, and V.P. Dudkevich, *Ferroelectrics* **192**, 313 (1997).