A dynamical model for fractal and compact growth in supercooled systems

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Abstract

A dynamical model that can exhibit both fractal percolation growth and compact circular growth is presented. At any given cluster size, the dimension of a cluster growing on a two-dimensional square lattice depends on the ratio between the rates of two probabilistic processes, namely (i) the aggregation of lattice sites into the growing cluster and (ii) the relaxation of lattice sites into those available for potential aggregation. The proposed model approaches the limit of two-dimensional invasion percolation if the aggregation process is much faster than the relaxation process, and it approaches Eden’s model for compact circular growth if the relaxation process is much faster than the aggregation process. Experimental examples of the fractal-growth regime include the percolation-like growth of bent-core smectics and calamitic smectics, where such fractal growth is attributed to the slow relaxation of molecules in a viscous supercooled medium.

1. Introduction

Fractal geometry has been highly successful in describing complex patterns formed in a wealth of growth processes, with examples ranging from dielectric breakdown, colloidal aggregations, biological systems, and geology [1–7]. Under nonequilibrium conditions, many soft matter systems exhibit growth of fractal-like structures, such as those observed for polymer networks, colloidal aggregates, and liquid crystal clusters [8–23]. For supercooled systems, a variety of patterns have been observed experimentally for the two-dimensional growth of liquid crystal phases out of an isotropic melt [24,25]. Apart from the typical growth of compact domains (figure 1(a)), irregular fractal percolation-like clusters (figure 1(b)) have been observed for a variety of liquid crystal phases [13–21] with a slow relaxation dynamics [26–33] and a large viscosity [30–32,34], where the corresponding mechanism of fractal growth remains to be understood.

To understand how a particular growth pattern is formed, computer growth models are often employed to simulate the underlying growth process. Well-known examples include the models of diffusion-limited aggregation (DLA) [35–39] and relevant diffusion-based models [40,41], cluster-cluster aggregation (CCA) [42] and percolation [43–52] for the growth of fractal-like clusters, as well as Eden’s model [53,54], including its noise-reduced [35,56], off-lattice [57] and anisotropy-corrected [58] variants, for the growth of compact-like structures. Such growth models, which do not involve any detailed molecular mechanism, provides a unified description of growth processes across different experimental systems. A common feature of such models is their probabilistic nature, where in a computational step each possible event (e.g. aggregation or diffusion) would occur at a finite probability. This, however, raises a conceptual question as to how such a finite probability is related to the corresponding rate (probability per unit time) of the event. If each computational step represents an infinitesimal time step, a finite probability would imply that the event occurs at an infinite rate, which is physically impossible.

In this paper, a dynamical growth model that unifies the concepts of rate and site-occupation probability for a two-dimensional square lattice is presented. Taking into account the aggregation rate of a growing cluster and

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the rate of a relaxation process in the medium, it is shown mathematically that each site-occupation probability corresponds to a conditional probability of the event but not the infinitesimal probability in the corresponding rate of occurrence. Numerical simulations show that the dynamical model reduces to either invasion percolation \cite{46} or Eden’s circular growth \cite{53,54} if one rate is much higher than the other.

This paper is organized as follows: In section 2, the dynamical growth model and the corresponding computational method are introduced. In section 3, simulation results of the growth model are presented and discussed. In section 4, experimental examples of both fractal percolation growth and compact circular growth are discussed in terms of their relaxation dynamics and viscosities, with a comparison of experimental values against the simulation results. In section 5, the findings of this study are summarized and the underlying physics of the model is discussed.

2. Theoretical model and computational method

Consider a single cluster growing on a two-dimensional square lattice, with each lattice site representing a micron-sized mesoscopic region of the supercooled system. At time $t = 0$, a nucleation site, which is defined as the very first site of the growing cluster, is placed at the centre of the lattice, and all other lattice sites are not yet available for potential aggregation into the growing cluster. At $t > 0$, the fraction of lattice sites available for potential aggregation relaxes from 0 towards 1 at some relaxation rate $f_r$. The cluster could start growing only after one of the four nearest neighbours of the nucleation site becomes available for aggregation. Growth of the cluster proceeds with repeated random aggregations of available nearest neighbours at some aggregation rate $f_a$, with only a single site aggregated at one time. As a first approximation, both $f_r$ and $f_a$ are assumed to be constant, without taking into account any possible temperature influence on the dynamics of aggregation or relaxation. Experimental results \cite{25,59–61} suggest that the assumed constancy of $f_a$ is valid if the depth of temperature quench is sufficiently large. Whenever a nearest neighbour becomes part of the cluster, the set of nearest neighbours would have to be updated. Using the software BENOIT1.3 (TruSoft International), the dimension of each simulated cluster was characterized via the box-dimension method \cite{1,2}:

$$N(d) \sim \frac{1}{d^D}$$

(1)

where $N(d)$ is the number of boxes of side length $d$ as occupied by the cluster. The same approach has been employed to characterize the fractal dimensions of experimental clusters \cite{13–18}.

The rates $f_r$ and $f_a$ can both be expressed as probabilities per unit time. For

$$f_r = \frac{dP_r}{dt},$$

(2)

the fraction of relaxed sites at any time $t$ is given by

$$P_{\text{sites}} = 1 - \exp(-f_r t).$$

(3)

Similarly, the aggregation rate for every available nearest neighbour is given by

$$f_a = \frac{dP_a}{dt},$$

(4)

$$N(d) \sim \frac{1}{d^D}$$

Figure 1. Growth textures of (a) the nematic phase of a calamitic liquid crystal and (b) the B2 phase of a bent-core liquid crystal, as observed via polarizing microscopy. The grey and white regions correspond to the growing phases in (a) and (b), respectively. The colour contrast between each growing phase and the supercooled isotropic melt (dark background) is a result of optical birefringence. The nematic phase in (a) exhibits compact circular growth with a Euclidean dimension of $D = 2$, and the B2 phase in (b) exhibits percolation growth with a fractal dimension of $D \sim 1.9$. 
where the overall aggregation rate is defined to be this aggregation rate multiplied by the number of available nearest neighbours. The morphology of the growing cluster at any given value of \( p_{\text{sites}} \) depends only on the ratio

\[
\nu \equiv \frac{f_a}{f_i} = \frac{dP_a}{dP_i},
\]

but not on the absolute values of \( f_a \) and \( f_i \), because the system’s evolution is driven solely by the infinitesimal probabilities \( dP_a \) and \( dP_i \).

Consider a total of \( N \) possible events of relaxation or aggregation in the system, with their infinitesimal probabilities of occurrence denoted as \( \{dP_1, dP_2, \ldots, dP_N\} \), respectively. The probability that any two or more events occur simultaneously is a product of infinitesimal quantities, and should therefore be neglected. It follows that, at any moment, there is either one single event or none occurring. Take event 1 as an example. The probability that only event 1 occurs in the system is also \( dP_1 \), as illustrated by the following equation:

\[
dP_1 \times (1 - dP_2) \times (1 - dP_3) \times \ldots \times (1 - dP_N) = dP_1
\]

where, for \( i \in [1, N] \), the term \( (1 - dP_i) \) is the probability that event \( i \) does not occur. The system’s dynamics can then be simulated by extracting all the single-event moments. By imposing the condition that a single event occurs, the conditional probability for the occurrence of event 1 is given by

\[
P_1 = \frac{dP_1}{\sum dP_i}
\]

where the denominator is the probability sum of all possible events over the whole system and also the probability that a single event occurs in the system. If event 1 occurs, the mean time interval between this single event and the previous one is given by

\[
\tau = \frac{dt}{\sum dP_i}
\]

The time \( t \) at which this event occurs is approximated as the sum of all \( \tau \)-intervals prior to the event. An arbitrary time scale can be chosen because the absolute values of time play no role in the morphological evolution of the growing cluster.

3. Simulation results

Simulations were carried out on a 1000 \( \times \) 1000 square lattice with a single nucleation site at the lattice centre. The values of \( \nu \) were chosen between two comparatively large and small values, \( \nu = 10^3 \) and \( \nu = 10 \), which correspond to relatively slow and fast relaxation within the medium, respectively. At a cluster size of 50 000 lattice sites, there already exists a significant difference in cluster morphology between the two cases. At \( \nu = 10^3 \), the cluster grows in a fractal percolation-like pattern (figure 2(a)) with its dimension saturating at \( D \sim 1.9 \) for increasing cluster size (figure 3(a)). Note that this is in practical agreement with the theoretical value of \( D = 91/48 \approx 1.8958 \) [50, 52]. At \( \nu = 10 \), the cluster grows in a compact circular fashion (figure 2(b)) with a cluster dimension of \( D = 2 \) (figure 3(b)).

Consider the percolation threshold \( p_c \approx 0.5927 \) [48] for a two-dimensional square lattice. At \( p_{\text{sites}} < p_c \), the relaxed sites of the medium do not form a long-range connectivity so that growth of the cluster would be inhibited at some point. When \( p_{\text{sites}} \) reaches \( p_c \), a long-range connectivity of those relaxed sites is established so that the cluster can grow continuously. This is demonstrated by the ‘explosive’ growth for large values of \( \nu \) (figure 4(a)). Such ‘explosive’ growth is not observed for small values of \( \nu \) (figure 4(b)). For \( \nu = 10^3 \), relaxation within the medium is sufficiently slow such that \( p_{\text{sites}} \) remains quasistatic around \( p_c \) for increasing cluster size (figure 3(a)). The fractal dimension thus saturates at \( D \sim 1.9 \). For \( \nu = 10 \), relaxation is relatively fast so that \( p_{\text{sites}} \) has already increased to values well above \( p_c \) at an early growth stage (figure 3(b)). The cluster dimension is thus \( D = 2 \) as observed for compact domains in two-dimensional space.
As mentioned above, the probability of any set of simultaneous events is a product of infinitesimal quantities and thus should be neglected. Therefore, fractal growth at $\nu = 10^5$ is a process of invasion percolation because the growing cluster ‘invades’ only one adjacent relaxed site at one time [48, 49]. For the same reason, circular growth at $\nu = 10$ and $p_{\text{sites}} \to 1$ is a growth process described by Eden’s model where only one adjacent site is chosen randomly for each growth step [53]. The compact-to-fractal crossover of growth morphologies from small to large values of $\nu$ was also investigated, with a series of simulated growth images presented in figure 5. It was found that the corresponding Euclidean-to-fractal dimensional crossover is continuous and exhibits scaling behaviour. For example, the crossover regime for a cluster size of 50 000 lattice sites scales as $D \sim \nu^{-0.016}$ (figure 6(a)). In any follow-up research, detailed numerical investigations on the cluster-size dependence of such scaling should be carried out. It was also found that the crossover regime generally shifts towards larger $\nu$ values for increasing cluster size, as illustrated in figure 6(b). This implies that, if the simulations can ideally be run for an indefinite amount of time, any growing cluster would eventually crossover from fractal percolation growth to compact circular growth after a certain cluster size is reached.

For varying $p_{\text{sites}}$, a previous study [47] also generated a crossover between compact growth and percolation growth on a two-dimensional square lattice. At $p_{\text{sites}} = 1$, however, the compact clusters were square-shaped, i.e. anisotropic. In every aggregation step, the nearest neighbours of the growing cluster were all aggregated deterministically, so that the resulting cluster morphology fully reflects the structural anisotropy imposed by the square lattice. In the present model, only a single nearest neighbour is picked randomly for each aggregation event. The probabilistic nature of this algorithm obscures the structural anisotropy imposed by the lattice, so that the compact growth patterns at $p_{\text{sites}} = 1$ appear to be isotropic and radially symmetric. Similarly, the lattice-imposed anisotropy in percolation clusters obtained from the present model is not obvious, because such anisotropy is obscured by the random spatial distribution of relaxed sites at $p_{\text{sites}} \to 1$.
4. Experimental examples

The theoretical investigation presented above was originally motivated by the observation of contrasting patterns for the growth of liquid crystal phases out of a supercooled amorphous melt (figure 1). As observed via polarising microscopy and digital-image acquisition (figure 7), standard nematic liquid crystals exhibit compact growth of circular domains (figure 1(a)), while the B2 phase of a bent-core liquid crystal and the smectic C phase...
of a calamitic liquid crystal grow in the form of irregular fractal-like structures [13–18, 21] (figure 1(b)) with an observed dimension very close to that of percolation clusters. Experiments suggest a correlation between relaxation slowdown [26–28] and fractal growth for bent-core smectics and calamitic smectics, with relaxation frequencies \( f_{r, \text{calamitic nematics}} \gg f_{r, \text{bent-core nematics}} \sim f_{r, \text{calamitic smectics}} \gg f_{r, \text{bent-core smectics}} \) and shear viscosities \( \eta_{r, \text{calamitic nematics}} \ll \eta_{r, \text{bent-core nematics}} \sim \eta_{r, \text{calamitic smectics}} \ll \eta_{r, \text{bent-core smectics}} \) for the various types of liquid crystals under consideration. The shear viscosity of a standard calamitic nematic phase is in the orders of 0.01 to 0.1 Pa·s [62, 63]. For such calamitic nematic phases, dielectric-spectroscopy experiments reveal that the relaxation frequencies of the molecular modes are in the orders of \( 10^7 \) to \( 10^8 \) Hz [64], which corresponds to relaxation times in the orders of 10 to 100 ns. Nematic phases of bent-core molecules typically exhibit a higher viscosity and a slower relaxation dynamics of molecules than their calamitic nematic counterparts, with a difference of one to two orders of magnitude. The viscosity, relaxation frequency and relaxation time of a bent-core nematic phase are in the orders of 1 to 10 Pa·s [34], \( 10^3 \) to \( 10^6 \) Hz [29] and 1 to 10\( \mu \)s, respectively, where these values are comparable to those for fluid smectic phases of calamitic molecules [30–32]. Smectic phases of bent-core liquid crystals, such as the fractal-growing B2 phase (figure 1(b)), typically exhibit a slower relaxation dynamics of molecules than their calamitic counterparts, with relaxation frequencies and relaxation times in the orders of \( 5 \times 10^3 \) to \( 5 \times 10^6 \) Hz [33] and 20 to 200\( \mu \)s, respectively. Thus the relaxation rate of a fractal-growing bent-core smectic phase is typically \( 10^3 \) to \( 10^4 \) times lower than that of a calamitic nematic phase. This order of

![Figure 6](image.png)

Figure 6. (a) Continuous Euclidean-to-fractal dimensional crossover from small to large values of \( \nu \) at a cluster size of 50 000 lattice sites, with \( D \sim \nu^{-0.016} \) in the crossover regime; (b) \( \rho_{\text{sites}} \) as a function of \( \nu \) for cluster sizes of 10 000 and 50 000 lattice sites, showing a shift of this profile towards larger \( \nu \) values for increasing cluster size. This implies that any growing cluster would eventually crossover from fractal percolation growth to compact circular growth after a certain cluster size is reached.
difference is in agreement with that of $f_r = f_a/\nu$ as predicted by the present model (figure 6(a)) if, as a first approximation, the differences in $f_a$ among the various types of liquid crystals could be neglected.

5. Conclusions and future work

A dynamical model that describes two-dimensional growth processes in a time-varying supercooled medium is presented. The model approaches the limit of two-dimensional invasion percolation if the aggregation rate $f_a$ of the growing cluster is much higher than the relaxation rate $f_r$ of the medium, and it approaches Eden’s model for compact circular growth if the relaxation rate $f_r$ is much higher than the aggregation rate $f_a$. The model finds applications in describing the growth behaviour of a variety of liquid crystal phases. Both numerical simulations and experimental examples suggest that the rate ratio $\nu = f_a/f_r$ for fractal percolation growth is at least three to four orders of magnitude larger than that for compact circular growth.

The model corresponds to the following physics of fractal growth: The saturation of observed fractal dimensions of experimental liquid crystal clusters at $D \sim 1.9$ for non-diffusive percolation, but not at $D \sim 1.7$ [2, 3] for diffusion-limited aggregation, suggests the followings: (1) Once a long-range connectivity of stationary relaxed regions is established, percolation overtakes diffusion-limited aggregation as the dominant growth mechanism; (2) Compared to percolation growth, diffusion-limited growth is too slow to be observed, due to the significant slowdown of molecular diffusion in a viscous supercooled medium. The relatively slow relaxation of molecules is attributed to the relatively high viscosity of the medium, in which it would generally take more time for the molecules to overcome viscous forces and for them to self-assemble into the more ordered growing phase. The irregularity of any percolation-like cluster is thought to be arising from the random inhomogeneity of a partially relaxed supercooled medium, where such inhomogeneity is represented by a random distribution of relaxed sites in the model.

For future work, the model could be generalized along the following directions: (1) Some liquid crystal phases exhibit anisotropic growth in the form of elongated domains [25, 60, 61]. For such systems, the model could be generalized to growth processes with anisotropic rates of aggregation. (2) For cases of isotropic growth, any growth process simulated on a square lattice must bear the unrealistic effect of lattice-imposed anisotropy [54, 55]. Potential solutions to this problem include the development of an off-lattice [57] or anisotropy-corrected [58] version of the model. (3) For cases of shallow temperature quench, the growth dynamics is typically limited by the diffusion of latent heat [25, 59–61, 65–70]. It would be worth taking into account the effects of thermal diffusion and the temperature dependence of aggregation rates.
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