Persistence, Poisoning, and Autocorrelations in Dilute Coarsening

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We calculate the exact autocorrelation exponent λ and persistence exponent θ, and also amplitudes, in the dilute limit of phase ordering for dimensions $d \geq 2$. In the Lifshitz-Slyozov-Wagner limit of conserved order parameter dynamics we find $\theta = \gamma_d \epsilon$, a universal constant times the volume fraction. For autocorrelations, $\lambda = d$ at intermediate times, with a late time crossover to $\lambda \geq d/2 + 2$. We also derive λ and θ for globally conserved dynamics and relate these to the $q \to \infty$–state Potts model and soap froths, proposing new poisoning exponents. [S0031-9007(97)04799-6]

While much has been learned about the coarsening kinetics that follows a temperature quench from a single-phase to a multiphase state [1], relatively little has been established for certain recently introduced dynamical exponents. It is accepted that the characteristic length scale of strongly correlated regions grows as a power law in time, $L \sim t^{1/z}$, with universal z [1]. Most nonconserved order parameter systems yield $z = 2$, and those with scalar conserved order parameter dynamics yield $z = 3$, *independent* of the system dimensionality d or of conserved quantities such as the volume fraction ϵ of the minority phase. Consequently, persistence [2–11] and autocorrelation [12–17] exponents, θ and λ, respectively, are being explored in the hope that they contribute to a characteristic set of universal exponents analogous to those of equilibrium criticality. However, fundamental questions remain about the universality of these new exponents, and even of the existence of power laws in the relevant quantities.

The persistence exponent θ, introduced in the experimental study of breath figures [2], is defined by the power-law decay, $P(t_1, t_2) \sim t_2^{-\theta}$, of the probability that a stochastic variable has not crossed some threshold—typically its mean—between the times t_1 and t_2 [3]. We consider the persistence of a *local* scalar order parameter $\phi(\mathbf{r}, t)$ (rescaled so that in equilibrium $\phi = \pm 1$), given by the fraction of the system that has not undergone phase change between t_1 and t_2 [4].

In $d = 1$, θ has been calculated exactly for the nonconserved q-state Potts model [5] and has been shown to be universal by renormalization group methods in the Ising ($q = 2$) case [6]. In higher dimensions, studies have focused on diffusion models [7], which exhibit nontrivial values of θ, and on related Gaussian approximations for nonconserved ordering kinetics [7,8]. For $d = 2$, these approximate results compare well with simulations [9,10] and with experiments on twisted nematic liquid crystals [11], an Ising analog. However, there have been no previous studies of persistence for conserved coarsening dynamics.

In phase-ordering systems the autocorrelation function, $A(t_1, t_2) \equiv \langle \phi(\mathbf{r}, t_1) \phi(\mathbf{r}, t_2) \rangle - \langle \phi \rangle^2$, decays asymptotically as $A(t_1, t_2) \sim [L(t_1)/L(t_2)]^\lambda$, which defines λ [1,12]. For nonconserved scalar coarsening, λ has been measured experimentally in $d = 2$ [13] and calculated in $d = 1$ [14]. Approximate calculations and numerical results have been obtained for various ϵ and d [15] in the case of *globally* conserved dynamics—i.e., a nonconserved order parameter (hence $z = 2$) subject to a constraining field that maintains the total volume fraction of each phase. *Locally* conserved dynamics has been studied numerically in two [16,17] and three [17] dimensions, and a formal asymptotic bound, $\lambda \geq d/2 + 2$, has been established by Yeung, Rao, and Desai (YRD) [16], but otherwise little theoretical progress has been made.

In this Letter, we study persistence and autocorrelations for both locally and globally conserved dynamics. We focus on the asymptotic late stage regime which follows a quench to a subcritical temperature $T < T_c$, in the limit of vanishing volume fraction, $\epsilon \to 0^+$. This is the limit of the classic Lifshitz-Slyozov-Wagner (LSW) theory, which firmly established $z = 3$ for dilute locally conserved coarsening [1,18]. We use the LSW theory in a similar spirit to obtain θ and λ. A summary of our results follows.

For locally conserved dynamics we compute θ for all $d \geq 2$ and demonstrate that (i) the persistence decays as a power law, $P(t_1, t_2) \sim (t_1/t_2)^\theta$, (ii) the exponent is a function of the volume fraction, going as

$$\theta = \gamma_d \epsilon \tag{1}$$

in the small ϵ limit, and (iii) γ_d is universal in that it does not depend on the surface tension, quench depth, temperature, or mobility, with $\gamma_2 \simeq 0.390\,08$, and $\gamma_3 \simeq 0.509\,45$. A large-$d$ expansion gives the asymptotic series

$$\gamma_d = \sqrt{3d/8\pi}\left[1 + \sum_{m=1}^{k} a_m d^{-m} + O(d^{-k-1}) \right], \tag{2}$$

which is quite accurate in $d = 2, 3$ when truncated at $k = 3$ [19,20]. We also compute the order $\epsilon^{3/2}$ corrections to (1) in $d = 3$.

For the autocorrelation function we find $\lambda = d$ as $\epsilon \to 0^+$, with explicit universal expressions for both the amplitude *and* logarithmic corrections. We also present a physical scaling argument that predicts a crossover to $\lambda \gtrsim d/2 + 2$ for any finite ϵ, after $t_2 \gtrsim \epsilon^{-3/d} t_1$, thus satisfying the YRD bound [16].

Next we consider globally conserved (GC) dynamics, again in the small ϵ limit, and find θ to have the same form (1), with a different universal γ_d^{GC}. In particular, $\gamma_2^{GC} \simeq 0.48797$ and $\gamma_3^{GC} \simeq 0.62450$. In large d

$$\gamma_d^{GC} = \sqrt{d/2\pi}\,[1 - \tfrac{1}{3}d^{-1} + \tfrac{43}{288}d^{-2} + O(d^{-3})], \quad (3)$$

which is highly accurate for all d [20]. The large d asymptote, $\theta \simeq 0.40\epsilon\sqrt{d}$ is similar to the approximate result $\theta \simeq 0.15\sqrt{d}$ obtained at $\epsilon = \tfrac{1}{2}$ [7] (where the GC dynamics is equivalent to nonconserved). The autocorrelation exponent is $\lambda = d$ [15], with no crossover expected at late times. We also find universal amplitudes *and* logarithmic corrections.

Finally we draw a connection between the above results for *persistence* in GC dynamics with *poisoning* (defined below) in soap froths. First, there is some evidence, mainly in $d = 2$, that soap froths have the same asymptotic dynamics as the nonconserved q-state Potts model in the $q \to \infty$ limit [21]. Second, the coarsening of the q-state Potts model and that of GC dynamics with $\epsilon = 1/q$ were shown to be equivalent as $q \to \infty$, within a Gaussian approximation scheme [15]. However, the details of the topological rearrangements may be different between Potts models and soap froths [21], and, further, while both Potts models and soap froths have vertices, GC systems do not.

Nevertheless, numerical studies found little difference in λ between the Potts and GC models [15], implying that they may lie in the same dynamic universality class. Also, the Potts persistence exponent, which measures the volume never visited by a wall, is given via the GC correspondence by $\theta = d/2$ as $q \to \infty$ [22], consistent with Potts simulations [9,23] and with experiments on $d = 2$ soap froths [24]. To further explore these analogies, we define a new set of *poisoning* exponents θ_Σ that give the decay of volume that has never been visited by any of a *set* of phases that occupy a *total* volume fraction Σ. This poisoning should provide a more delicate test of the underlying dynamics than autocorrelations or persistence and may be directly explored via simulations of Potts models and experiments on foams [25]. By use of the GC correspondence we obtain, for $\Sigma \ll 1$,

$$\theta_\Sigma = \gamma_d^{GC} \Sigma. \quad (4)$$

We begin with the LSW theory, which applies to widely separated drops of minority phase as $\epsilon \to 0^+$. This theory provides the only solution of a phase-ordering system in $d > 1$ with topological defects—in this case with domain walls. Drops of radius R evolve according

to $R^2\dot{R} = \alpha_d(R/R_c - 1)$, where α_d is a nonuniversal constant [1] and the dot indicates a time derivative. Here $R_c = (\tfrac{4}{9}\alpha_d t)^{1/3} \sim L$ is the critical radius, where drops shrink for $R < R_c$ and grow for $R > R_c$. The density $n(R, t)$ of droplets of size R at time t obeys a continuity equation $\dot{n} = -\partial_R(\dot{R}n)$, which leads to a scaling solution $n_d(R, t) = R_c(t)^{-d-1} f_d(R/R_c)$, with

$$f_d(x) = \frac{\epsilon F_d x^2 \exp[-d/(3 - 2x)]}{(3 + x)^{1+4d/9}(3 - 2x)^{2+5d/9}}, \quad (5)$$

and $f_d = 0$ for $x \geq 3/2$ [1,26,27]. The normalization constant F_d is determined by the volume fraction of the minority phase, $\epsilon = V_d \int_0^\infty dx\, f_d(x)\,x^d$, where $V_d \equiv \pi^{d/2}/\Gamma(1 + \tfrac{1}{2}d)$ is the unit d-sphere volume. This gives $F_2 \simeq 37.752$ and $F_3 \simeq 186.13$, and the large-d expansion $V_d F_d = e^d 2^{1/2+8d/9} \times \sqrt{27d/\pi}\,[1 + \sum_{m=1}^k a_m d^{-m} + O(d^{-k-1})]$ [19]. The total number density of drops can be shown to be $n(t) = \epsilon F_d/(4d3^d e^{d/3})[R_c(t)]^{-d}$.

The droplet growth equation can be written in terms of the scaled size $x \equiv R/R_c(t)$ as $3tx^2\dot{x} = -(x + 3)(x - \tfrac{3}{2})^2$; where $\dot{x} < 0$ for all $x \geq 0$. This may be integrated to give the trajectory

$$t_f = t(1 + \tfrac{1}{3}x)^{4/3}(1 - \tfrac{2}{3}x)^{5/3} \exp[2x/(3 - 2x)], \quad (6)$$

which is parametrized by the time of complete evaporation t_f. From (6), we find an expansion for $t_1 \ll t_2 \leq t_f$ which will prove to be useful:

$$x(t_1) = \tfrac{3}{2}[1 - \delta(t_1/t_2) + \cdots], \quad (7)$$

where $\delta(t_1/t_2) = 1/[\ln(t_2/t_1) + \tfrac{5}{3}\ln\ln(t_2/t_1)]$. This leading correction to $x(t_1)$, related to the essential singularity in $f_d(x)$ at $x = \tfrac{3}{2}$, is *universal* and independent of $x(t_2)$.

Using these LSW results, we can calculate $P^<(t_1, t_2)$, the persistent or unpoisoned volume fraction of minority phase, and $P^>(t_1, t_2)$, that of the majority phase. The total persistent volume $P = P^> + P^<$ will decay with the slower of the two unpoisoned fractions.

Only droplets that have survived to time t_2 contribute to $P^<$, the unpoisoned *minority* phase, and their density decays as $n \sim R_2^{-d} \sim t_2^{-d/3}$, using the notation $R_i \equiv R_c(t_i)$. The density of droplets that have survived and are *smaller* at t_2 than they were at t_1 is

$$R_2^{-d} \int_0^{x_1(t_1/t_2)^{1/3}} dx\, f_d(x) \sim R_2^{-d-3} \sim t_2^{-d/3-1}, \quad (8)$$

where we have used the small x behavior $f_d(x) \sim x^2$ for $t_2 \gg t_1$ and $R_c \sim t^{1/3}$. Hence, droplets that have shrunk comprise a vanishing fraction of those surviving at t_2, so that no surviving droplets have been poisoned at time $t_2 \gg t_1$. Consequently, $P^<$ is asymptotically the number density $n(t_2)$ times the volume at the initial time t_1 of those largest droplets, $V_d[x(t_1)R_1]^d$. Using $x(t_1)$ from (7),

$$P^<(t_1, t_2) = \tfrac{1}{4}\epsilon B_d(t_1/t_2)^{d/3}[1 - d\delta(t_1/t_2) + \cdots], \quad (9)$$

where $B_d = F_d V_d/(2^d d e^{d/3})$, with $B_2 \simeq 7.6115$ and $B_3 \simeq 11.951$, *and* the leading logarithmic corrections $\delta(t_1/t_2)$ are universal.

The *majority* phase, with unpoisoned volume fraction $P^>$, can only be poisoned by growing drops, i.e., those with $R > R_c$. Since the drop positions are uncorrelated in the dilute limit [18]—the key feature which makes the LSW theory soluble—it follows that the unpoisoned regions must be uncorrelated as well, leading to

$$\partial_t P^>(t_1, t) = -\dot{v}(t) P^>(t_1, t),\qquad(10)$$

where $\dot{v}(t)$ is the rate of encroachment by the minority phase. From the LSW theory we have

$$\dot{v}(t) = \int_{R_c}^{R_{max}} dR \; V_d R^d \partial_t n(R, t) = V_d R_c^d n(R_c, t) \dot{R}_c,\qquad(11)$$

where the second equality comes from mass conservation of drops larger than $R_c(t)$, i.e., $\partial_t \int_1^{3/2} dx \, x^d f_d(x) = 0$. Using $R_c \propto t^{1/3}$, we obtain $\dot{v} = V_d f_d(1)/(3t) \equiv \epsilon \gamma_d/t$ where

$$\gamma_d = F_d V_d/(4^{1+4d/9} 3 e^d)\qquad(12)$$

is a universal constant. Combining (10) and the initial condition, $P^>(t_1, t_1) = 1 - \epsilon$, we find

$$P^>(t_1, t_2) = (1 - \epsilon)[t_1/t_2]^{\gamma_d \epsilon},\qquad(13)$$

so $P^>$ indeed decays as a power law. Remarkably, this result is valid for all $t_2 > t_1$ in the scaling regime, not just when $t_2 \gg t_1$. As expected, $P^>$ decays slower than $P^<$, and so $P(t_1, t_2) \sim P^>(t_1, t_2)$, leading to Eq. (1) for θ.

In order to derive λ from the persistence, it is convenient to change field variables to $\psi = (\phi + 1)/2$ (with minority phase $\psi = 1$ and majority $\psi = 0$), giving $\langle \psi \rangle = \epsilon$ and the autocorrelation function $A(t_1, t_2) = 4[\langle \psi(\mathbf{r}, t_1) \psi(\mathbf{r}, t_2) \rangle - \epsilon^2]$. The two-time average is then the probability of finding a given point inside minority droplets at both t_1 and t_2. The contribution from unpoisoned regions is exactly $P^<(t_1, t_2)$, whereas poisoned regions that find themselves in a droplet again at t_2 contribute $[\epsilon - P^<(t_1, t_2)]^2$. To leading order in ϵ,

$$A(t_1, t_2) = \epsilon B_d (R_1/R_2)^d [1 - d\delta(t_1/t_2) + \cdots],\quad(14)$$

giving $\lambda = d$ in the dilute limit (this was noted before in the GC case [15]). This exponent depends solely on the existence of a scaling distribution of uncorrelated drops; in the LSW limit the details of the drop distribution and evolution serve only to determine the universal amplitude and leading logarithmic corrections.

We expect (14) to hold for R_2 much less than the drop separation at t_1, $\tilde{R}_1 \sim \epsilon^{-1/d} R_1$. For $\epsilon \to 0^+$ this is forever. Below we discuss $\epsilon > 0$, where the drop separation is finite and correlations must be considered.

We turn now to globally conserved dynamics, where the analog of LSW theory was given by Sire and Majumdar [15]. Droplet growth follows $\dot{R} = \alpha_d[1/R_c(t) - 1/R]$, where $R_c(t)$ represents a time-dependent applied field tuned to maintain the volume fraction ϵ. Combining the

droplet growth with the continuity equation, as in LSW, we find a scaling solution for the droplet density, $n(R, t) = R_c^{-d-1} f^{GC}(R/R_c)$, when $R_c(t) = (\frac{1}{2}\alpha_d t)^{1/2}$, with the distribution

$$f_d^{GC}(x) = \epsilon F_d^{GC} x(2 - x)^{-d-2} \exp[-2d/(2 - x)]\quad(15)$$

for $x < 2$, and $f_d = 0$ otherwise. The normalization condition $\epsilon = V_d \int_0^\infty dx \, x^d f_d^{GC}(x)$, determines F_d^{GC}, with $F_2^{GC} \simeq 16.961$ and $F_3^{GC} \simeq 120.29$. A large-d expansion yields the excellent approximation $V_d F_d^{GC} = e^{2d}\sqrt{2d/\pi}[1 - \frac{1}{3}d^{-1} + \frac{43}{288}d^{-2} - \frac{1033}{25920}d^{-3} + O(d^{-4})]$. The droplet density is $n^{GC}(t) = \epsilon F_d^{GC}(2e)^{-d}/d$ [20]. Integrating the scaled growth equation, $2tx\dot{x} = -(2 - x)^2$, gives the trajectory $t_f = t(1 - \frac{1}{2}x)^2 \exp[2x/(2 - x)]$. Hence, drops surviving to time $t_2 \gg t_1$ have

$$x(t_1) = 2[1 - \delta^{GC}(t_1/t_2) + \cdots],\qquad(16)$$

where the leading correction is $\delta^{GC}(t_1/t_2) = 2/[\ln(t_2/t_1) + 2 \ln\ln(t_2/t_1)]$. The density of drops that are smaller at t_2 than at t_1 decays as R_2^{-d-2}, so these are again negligible asymptotically. Consequently, the autocorrelation function $A(t_1, t_2) = 4P^<(t_1, t_2)$ is

$$A(t_1, t_2) = \epsilon B_d^{GC}(R_1/R_2)^d [1 - d\delta^{GC}(t_1/t_2) + \cdots],\qquad(17)$$

with $B_d^{GC} = V_d F_d^{GC}/(de^d)$, giving $B_2^{GC} \simeq 3.6057$, $B_3^{GC} \simeq 8.3623$. The leading logarithmic corrections in $\delta^{GC}(t_1/t_2)$ are universal.

The calculation of majority poisoning and persistence goes through the same as before, using (11) and (10), with the result $\theta = \gamma_d^{GC}\epsilon$. The different growth exponent $R_c \sim t^{1/2}$ gives $\gamma_d^{GC} = V_d f_d^{GC}(1)/2 = V_d F_d^{GC} e^{-2d}/2$. The result is (3).

Up to this point, calculations of, say, $f_d(x)$ have been for the leading $O(\epsilon)$ term, for which the drops may be regarded as uncorrelated. Higher order effects such as droplet collisions and diffusion-mediated interactions will lead to correlations in the drop sizes and positions. However, screening of the diffusion field has been shown in $d = 3$ to contribute $O(\epsilon^{3/2})$ corrections for *uncorrelated* drops [27–29], which is thus believed to represent the leading correction to LSW theory, with correlations coming in only at $O(\epsilon^2)$ [28]. Note that with GC dynamics there is no diffusion field, hence the leading corrections, due to collisions, are expected to be of $O(\epsilon^2)$.

With our existing machinery, then, we can compute the leading corrections to the LSW exponents. Since drops are uncorrelated to $O(\epsilon^{3/2})$, $\lambda = d$ remains unchanged. However, the distribution function is $\tilde{f}_3(x) = f_3(x)[1 + \epsilon^{1/2}\{G_3 + g_3(x)\}] + O(\epsilon^2)$, with

$$g_3(x) = b_0 \left[2\ln\left(\frac{3 + x}{\frac{3}{2} - x}\right) + \frac{14}{x + 3} + \frac{64x - 87}{4(3 - 2x)^2} \right],\qquad(18)$$

where $b_0 \equiv \frac{1}{9}\sqrt{\pi F_3/e} \simeq 1.6297$, and we have maintained $x \equiv R/R_c$ [20]. The normalization condition

determines $G_3 \simeq -3.4047$. This leads, via (11), to the persistence exponent $\theta \simeq 0.50945\epsilon - 0.14969\epsilon^{3/2} + O(\epsilon^2)$.

The leading correction to λ for $\epsilon > 0$ is of a different nature. At sufficiently late times t_2, λ becomes strongly dependent on correlation effects [20]. This occurs when drops grow to be larger than their earlier spacing, $R_2 \gtrsim \tilde{R}_1 \sim \epsilon^{-1/d} R_1$. Then the autocorrelation function is no longer given by the unpoisoned minority volume fraction, since each drop at t_2 covers *many* drops from t_1. As a result, the decay of autocorrelations becomes dominated by the fluctuations in drop density at t_1, which are described by the correlations. To see this, we use the small-k behavior of the structure, $S(k,t) \equiv \langle \psi_k \psi_{-k} \rangle \sim R_c^{d+4} k^4$ [30,31]. At time t_1, the fluctuations δV_ψ in the volume covered by $\psi = 1$ within an region of size R_2 is (up to numerical factors) $\delta V_\psi^2 \sim \int d^d k \int_0^{R_2} d^d r \int_0^{R_2} d^d r' e^{-ikr} S(k,t_1) \sim R_2^{2d} \int_0^{1/R_2} d^d k S(k,t_1)$, thus $\delta V_\psi \sim \pm R_1^{(d+4)/2} R_2^{(d-4)/2}$. In the case that the drops at t_2 coincide with positive fluctuations in the drop density at t_1, this volume δV_ψ will be contributed to $A(t_1,t_2)$ for every drop at t_2, of number density $1/R_2^d$. This gives the autocorrelation decay $A(t_1,t_2) \sim (R_1/R_2)^{(d+4)/2}$, which saturates the YRD bound [16]. In the case of weaker correlations between drops at t_2 and fluctuations in drop density at t_1, the autocorrelations decay faster. With both cases, we recover the YRD bound $\lambda \geq d/2 + 2$. Comparison with the $\epsilon \to 0^+$ result $\lambda = d$ shows that, at least for $d < 4$, the asymptotic decay of autocorrelations becomes faster when correlations suppress fluctuations in the drop density at large length scales.

In summary, we have demonstrated the existence of power laws in the autocorrelations, persistence, and poisoning of LSW and GC systems for $T < T_c$ coarsening, and have calculated the exact asymptotic amplitudes and exponents. Our results are universal for isotropic systems such as polymer blends (before hydrodynamic regimes). While the exponent θ is small in the dilute regime, it should be measurable since (13) holds for all $t_2 > t_1$.

In the future, we hope that the poisoning exponents θ_Σ are measured for foams and large-q Potts models as a sensitive probe of their equivalence, and compared with (4). It would also be worthwhile to extend the independent interval approximation [7] to the dilute globally conserved case, to further explore the common $\gamma_d \sim \sqrt{d}$ asymptote. Finally, our late-time crossover and logarithmic corrections in the autocorrelations may survive in larger filling fractions. Studying λ and its crossover as a function of ϵ may resolve the current ambiguities [16,17] about autocorrelation decay in conserved systems.

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