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Nucleation, Growth, and Scaling in Slow Combustion

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We study the nucleation and growth of flame fronts in slow combustion. This is modeled by a set of reaction-diffusion equations for the temperature field, coupled to a background of reactants and augmented by a term describing random temperature fluctuations for ignition. We establish connections between this model and the classical theories of nucleation and growth of droplets from a metastable phase. Our results are in good agreement with theoretical predictions.

KEY WORDS: Nucleation; reaction-diffusion systems; flame fronts.

I. INTRODUCTION

The kinetic process by which first-order phase transitions take place is an important subject of longstanding experimental and theoretical interest.⁽¹⁾ Nucleation is the most common of first-order transitions, and remains of a great deal of interest.⁽²⁻⁵⁾ There are two fundamentally different cases, homogeneous and heterogeneous nucleation. Homogeneous nucleation is an intrinsic process where embryos of a stable phase emerge from a matrix of a metastable parent phase due to spontaneous thermodynamic fluctuations. Droplets larger than a critical size will grow while smaller ones decay back to the metastable phase.⁽⁶⁾ More commonplace in nature is the process of heterogeneous nucleation. There, impurities or inhomogeneities catalyze a transition by making growth energetically favorable.

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Here we show that the concepts of nucleation and growth can be usefully applied to understand some aspects of slow combustion. We use a phase-field model of two coupled reaction-diffusion equations to study the nucleation and growth of combustion centers in two-dimensional systems. Such continuum reaction-diffusion equations have been used extensively in physics, chemistry, biology and engineering to describe a wide range of phenomena from pattern formation to combustion. However, the connection of reaction-diffusion equations to nucleation and interface growth has received little attention.

In a recent study of slow combustion in disordered media, Provatas *et al.*^(7, 8) showed that flame fronts exhibit a percolation transition, consistent with mean field theory, and that the kinetic roughening of the reaction front is consistent with the Kardar–Parisi–Zhang (KPZ)⁽⁹⁾ universality class. A very recent experimental study⁽¹⁰⁾ of slow combustion of paper indeed demonstrates that the roughening exponents obtained are compatible with the KPZ equation. In this paper we make a further connection between slow combustion started by spontaneous fluctuations, and the classical theory of the nucleation⁽⁶⁾ and growth of droplets from a metastable phase.

II. THE MODEL

We generalize the model of Provatas *et al.* by including an uncorrelated Gaussian noise source $\eta(\mathbf{x}, t)$, as a function of position $\mathbf{x}$ and time t , i.e., $\langle \eta(\mathbf{x}, t) \rangle = 0$ and $\langle \eta(\mathbf{x}, t) \eta(\mathbf{x}', t') \rangle = 2\varepsilon \delta(\mathbf{x} - \mathbf{x}') \delta(t - t')$, where the angular brackets denote an average, and ε is the intensity of the noise. The model then consists of equations of motion for the temperature field $T(\mathbf{x}, t)$ and the local concentration of reactants $C(\mathbf{x}, t)$. The temperature satisfies

$$\frac{\partial T(\mathbf{x}, t)}{\partial t} = D \nabla^2 T - \Gamma [T - T_0] + R(T, C) + \eta \quad (1)$$

The first term on the right-hand-side accounts for thermal diffusion, with diffusion constant D , the second term gives Newtonian cooling due to coupling with a heat bath of background temperature T_0 , with rate constant Γ , and the third term $R(T, C)$ is the exothermic reaction rate as a function of temperature and concentration of reactants. The concentration satisfies $\partial C(\mathbf{x}, t)/\partial t = -\lambda_1 R(T, C)$, and the reaction rate obeys

$$R(T, C) = \lambda_2 T^{3/2} e^{-A/T} C \quad (2)$$

where $\lambda_{1,2}$ are constants, A is the Arrhenius energy barrier, and Boltzmann's constant has been set to unity. Note that while the dynamics

of the process is controlled by the activation term $e^{-A/T}$, the scale for energy is set by $T^{3/2}$. We choose the same values for the constants as those used in ref. 8, which are approximately those for the combustion of wood in air: In physical units $D = 1 \text{ m}^2 \text{ s}^{-1}$, $\Gamma = 0.05 \text{ s}^{-1}$, $T_0 = 0.1 \text{ K}$, $A = 500 \text{ K}$, and the specific heat of wood, $c_p = 5 \text{ J g}^{-1} \text{ K}^{-1}$ (entering through $\lambda_2 = 8$). Length is measured in units of the reactant size, and time in units of those for the reaction to take place, λ_2/λ_1 . The low background temperature is chosen to permit good numerical accuracy for systems of moderate size, i.e., low T_0 ensures that diffusion fields remain short ranged as compared to the lattice size. The parameter ε was varied in the range $2 \times 10^{-7} - 11 \times 10^{-7}$: $\varepsilon = 2 \times 10^{-7}$ corresponds to slow, $\varepsilon = 5 \times 10^{-7}$ to medium, and $\varepsilon = 11 \times 10^{-7}$ to fast nucleation rate. Physically, ε cannot be related to temperature but it should be understood as the intensity of all external sources of noise.

We integrate these equations using Euler difference rules in space and time, with the smallest length $\Delta x = 1$, and time $\Delta t = 0.01$. Reactant units are randomly dispersed across the grid points with a probability c so that $C(\mathbf{x}, t) = 0$ for an unoccupied site and $C(\mathbf{x}, t) = 1$ for an occupied site, and evidently $0 \leq c \leq 1$. Figure 1 shows typical results for a two-dimensional system of size 256×256 , with periodic boundary conditions. The morphology of burnt and unburnt zones in these figures is strikingly similar to that for the nucleation and growth of crystallites from a supercooled melt,⁽¹⁾

Fig. 1. A snapshot of the process with medium nucleation rate, where $\varepsilon = 4.9 \times 10^{-7}$. Gray scale images temperature, where black is the hottest region. a) A system with uniform background ($c = 1$) at time $t = 16$ and b) a disordered system with $c = 0.5$ (which is still well above the percolation threshold $c^* \approx 0.2$ for $\varepsilon = 0$) at time $t = 20$. With quenched disorder ($c = 0.5$) domains burn at significantly lower temperature, and with more ragged boundaries, due to the lower concentration of reactants.

which is the motivation for our approach. We now briefly review the classical theories of nucleation and growth for such systems where a conservation law does not control the growth process.

III. THEORETICAL BACKGROUND

A. Classical Theory of Nucleation

The classical theory of nucleation⁽⁶⁾ has two main results: A description of the critical droplet, and a rate equation for the growth of clusters. The extra free energy due to a droplet of stable phase, in a metastable background, is $\Delta F = -V \Delta f + A\sigma$, where V is the volume of the droplet, A is its area, σ is surface tension, and Δf is the difference in the bulk free energy densities between the metastable and stable phases. The critical radius ρ^* of a droplet is obtained from through minimization: $\partial \Delta F / \partial \rho^* = 0$. In two dimensions, this gives $\rho^* = \sigma / \Delta f$. For the metastable phase to decay, droplets with $\rho > \rho^*$ nucleate and grow; droplets smaller than the critical radius shrink and disappear. The rate-limiting process is the critical droplet, with energy barrier $\Delta F(\rho^*)$, whose probability of occurrence is proportional to $\exp[-\Delta F(\rho^*)/T]$.

B. Kolmogorov–Avrami–Mehl–Johnson Theory

After nucleation has occurred, the subsequent growth of droplets is often well described by the phenomenological Kolmogorov–Avrami–Mehl–Johnson (KAMJ)^(11–13) model. This treatment describes many solid–solid and liquid–solid transformations, provided long-range interactions between droplets (which can be due to elastic effects, or diffusion fields) are of negligible importance. The KAMJ theory assumes that nucleation is a non-correlated random process with isotropic droplet growth occurring at constant velocity, where the critical radius is infinitesimal, and growth ceases when growing droplets impinge upon each other. As its basic result, the KAMJ description gives a functional form for the volume fraction of the transformed material:

$$X(t) = 1 - \exp \left[-\frac{V}{d+1} I v^d (t - t_0)^{d+1} \right] \quad (3)$$

for homogeneous (spontaneous) nucleation, and

$$X(t) = 1 - \exp[-V \alpha v^d (t - t_0)^d] \quad (4)$$

for heterogeneous (impurity induced) nucleation. In the above, v is the growth velocity, I is the nucleation rate and α is the density of embryos with $\rho > \rho^*$ present in the beginning of the process. Homogeneous and heterogeneous-nucleation are conveniently distinguished by the Avrami exponent, which are $d+1$, and d , respectively. The waiting time accounts for initial transients.

In the KAMJ description there are two intrinsic length scales present in the system. The first one is the critical radius and the other is found by simple dimensional analysis. As seen from Eqs. (3) and (4), the process is characterized by two variables: the nucleation rate and the growth velocity. Using them, the characteristic length for homogeneous nucleation, Eq. (3), can be written as $\xi = (v/I)^{1/(d+1)}$ and the characteristic time scale as $\tau = (Iv^d)^{-1/(d+1)}$. In practice, it is convenient to scale the time by the half-time of the transformation, $t_{1/2}$, since it is an easily accessible quantity both experimentally and computationally, and it can be used as a measure.⁽²⁾ In the limit where $\xi \gg \rho^*$ there is only one length scale present.⁽¹⁵⁾ During nucleation and growth this is the case up to the point when a connected cluster spans the system. As a consequence of this, scaling of $X(t)$ is expected, as we will show below. In the case of heterogeneous nucleation, Eq. (4), nucleation enters as an initial condition, thus being time independent. The system size is the only relevant length scale defining the characteristic time scale together with the velocity. Again, $t_{1/2}$ provides a convenient measure.

The apparent simplicity of the KAMJ description is due to the fact that it incorporates no correlations. For cases where such correlations are minimal (as for the case considered herein), it has been quite successful in describing experimental data,^(2,3) and theoretical generalizations can be readily made.^(5,14,15) The KAMJ description can be used in calculations of kinetic parameters and activation energies, and it provides information about the nature of the phase transition, i.e., if the process is diffusion or reaction (interface) controlled and if the process is influenced by inhomogeneities. Unfortunately, the basic KAMJ theory provides no information about the structural changes occurring during the phase transformation. Based on the same assumptions, Sekimoto⁽¹⁶⁾ derived exact analytical expressions for two-phase correlation functions when $\xi \gg \rho^*$. Fourier transforming Sekimoto's result for the two-point equal-time correlation function gives the structure factor:

$$S(\mathbf{k}, t) = \int d\mathbf{r} [C_2(\mathbf{x}, t; \mathbf{x} + \mathbf{r}, t) - C_1^2(\mathbf{x}, t)] e^{i\mathbf{k} \cdot \mathbf{r}} \quad (5)$$

where $C_1(\mathbf{x}, t)$ is the one-point correlation function equal to the KAMJ expression for transformed volume given in Eqs. (3) and (4). The two-point

correlation function C_2 is $C_2(\mathbf{x}, t; \mathbf{x} + \mathbf{r}, t) = C_1^2(\mathbf{x}, t) \exp[Iv^2\Psi(y)]$, where

$$\Psi(y) = \frac{2}{3} \left[\arccos(y) - 2y \sqrt{1-y^2} + y^3 \ln \left(\frac{1 + \sqrt{1-y^2}}{y} \right) \right]$$

for $y \leq 1$, and $\Psi(y) = 0$ for $y > 1$. The variable y is the normalized distance between two points. Substituting these into Eq. (5) gives

$$S(\mathbf{k}, t) = A \int_0^1 dy [e^{Iv^2 t'^3 \Psi(y)} - 1] y J_0(aky) \quad (6)$$

where J_0 is the Bessel function of the zeroth kind, $A = 2\pi a^2 e^{-(2/3)\pi I v^2 t'^3}$, and $a = 2vt'$ with $t' = t - t_0$.

C. Nucleation of Flame Fronts

We expect these theories to give a reasonable description of the growth of the flame fronts. In fact the agreement is much more impressive than we had anticipated. Of course, for combustion, the picture of nucleation and growth must be modified or re-interpreted in straightforward ways. For example, no shrinking of droplets, which herein correspond to burned patches, is possible. Also, instead of temperature in the Boltzmann probability weight, a quantity proportional to the intensity of noise sources ε must appear. Furthermore, the surface tension, evident in the fact that the burned patches are round, must have its origin in the dynamical description. Indeed, in the absence of nucleation it has been shown in refs. 7 and 8 that the flame front roughens according to the KPZ interface equation.⁽¹⁷⁾

For example, the critical radius, corresponding to the unstable stationary solution of the equation of motion for a droplet, can be found as follows. We follow refs. 4 and 17, and write the dynamical equation in circular coordinates as

$$\frac{\partial \rho}{\partial t} = \frac{D}{\rho^2} \frac{\partial^2 \rho}{\partial \theta^2} - \frac{D}{\rho} + \frac{v}{2} \left[2 + \left(\frac{1}{\rho} \frac{\partial \rho}{\partial \theta} \right)^2 \right] + \frac{1}{\sqrt{\rho}} \mu(\theta, t) \quad (7)$$

where μ is a linear combination of an effective noise due to the random reactant, and the additive noise η . In the limit of a flat front Eq. (7) corresponds to the KPZ equation. As explicitly shown in ref. 8, the equation of motion for the flame front can be mapped onto the KPZ equation.

To find the critical radius we need the velocity of the flame front. Using the flat front mapping,⁽⁸⁾ the velocity of the front is given as

$$v = (\Gamma A - \hat{\lambda}c)/\sigma \quad (8)$$

The constants A , $\hat{\lambda}$ and σ depend functionally on the temperature $T_m(x)$ that solves Eqs. (1) and (2) in the mean field limit. Their precise forms, which are not needed for our present purposes, are given in ref. 8: Physically, the constant A is proportional to the heat loss in the mean field limit, $\hat{\lambda}$ is proportional to the heat produced at the interface in the mean field limit, and σ is analogous to surface tension. To find an expression for the critical radius, we Fourier expand⁽⁴⁾ ρ and μ as $\rho(\theta, t) = \sum_n \rho_n(t) e^{in\theta}$, and $\mu(\theta, t) = \sum_n \mu_n(t) e^{in\theta}$. Substituting them into Eq. (7) together with the velocity, Eq. (8), gives $\rho^* = D\sigma/(\Gamma A - \hat{\lambda}c)$ for the lowest order estimate for the critical radius of a radial flame front. Simple dimensional analysis leads to the same result. When $\Gamma A \rightarrow \hat{\lambda}c$, $v \rightarrow 0$, i.e., the heat lost to thermal dissipation exactly balances that due to thermal reaction, and the critical radius goes to infinity. Unfortunately, the numerical window in which the critical radius changes appreciably is narrow, and close to c^* . Hence, although our numerical work reported below is consistent with the above analysis, it does not permit a quantitative test of our estimate of ρ^* . Finally, we would like to point out that for this analysis to be valid, the critical radius must be larger than the typical width of the front in order to satisfy the sharp interface approximation.

IV. NUMERICAL RESULTS

In our simulations we have typically used lattices of size 256×256 with periodic boundary conditions, averaging over 1000 sets of initial conditions. Heterogeneous nucleation is modeled via initial fluctuations at $t=0$, and homogeneous nucleation as time dependent Gaussian fluctuations through η . First, we compare our results for the fraction of burnt reactant product $X(t)$ to the KAMJ theory. We have simulated systems with various noise intensities with uniform ($c=1$) and disordered ($c=0.3$) reactant concentrations to see the applicability of the KAMJ theory in relation to this model. In both uniform and disordered cases the simulations are in excellent agreement with the theory as seen in Fig. 2. The case of homogeneous nucleation fits the Avrami exponent 3, while the heterogeneous case gives 2, as expected. In the scaled plots we have discarded the waiting time t_0 (Eqs. (3) and (4)) since it is due to lattice effects and the fairly small system size, and therefore does not represent a true physical time scale.

Fig. 2. Fraction burned, $t/t_{1/2}$ for various nucleation rates in homogeneous and heterogeneous nucleation in uniform and disordered systems. The inset shows the same data sets without scaling by $t_{1/2}$. The data is indistinguishable from the KAMJ theory (solid lines).

Next, we will focus on the structure factor in the case of uniform homogeneous nucleation. Here, $S(\mathbf{k}, t)$ corresponds to correlations in reactant concentrations, i.e., between burnt zones. In the cases of both high and low noise, we find a quantitative agreement between the theory and simulations at late times, as seen Fig. 3. In order to use the theoretical prediction, Eq. (6), we measured the growth velocity of the radius of individual nucleation centers for various concentrations, and found it to be in agreement with previous results,^(7, 8) i.e., $R(t) \sim t$.

As seen from Fig. 3, the theoretical prediction underestimates the rate of phase transformation during the early stages, but is in good agreement at later times. This is because, for early times, contributions to the structure factor from the bulk interior of droplets, and from the diffuse surface width of droplets are comparable. For later times, the surface contributions (not considered in the KAMJ and Sekimoto theories) are negligible.

For a uniform background there are very pronounced wiggles present, as can be seen from Fig. 3b. Their origin can be traced to the presence of the Bessel function in Eq. (6). The oscillations are due to the spherical

Fig. 3. (a) The structure factor vs. k for various time steps at a low noise intensity (see Fig. 2) for a uniform system ($c = 1$). (b) The structure factor vs. k with a medium noise intensity (see Fig. 2) for a disordered system ($c = 0.3$). The symbols represent the data from the simulations and the lines display the theoretical prediction obtained by integrating Eq. (6). The inset ($c = 1$) shows the structure factor vs. time at fixed k ; circles, $k = 0.1225$, and triangles, $k = 0.1975$. The long-dashed line shows Porod's law, which describes large- k correlations of randomly-oriented interfaces of negligible width.

shape of the burnt zones at early stages of the process when the growing regions have not yet merged with each other. The absence of wiggles in the case of quenched disorder, Fig. 3b, is simply due to the fact that the disorder affects the spreading of the temperature field resulting in kinetic roughening. This is also clearly visible in Fig. 1. In Figs. 3a and 3b we have also compared the numerical results to Porod's law, $S(k, t) \sim 1/k^{d+1}$ at large k .⁽¹⁾ This should describe a locally flat and thin interface, and indeed we find excellent agreement for large k .

To confirm that the origin of the discrepancies between the theory and our simulations of the structure factor are due to the interface width, we also compared the results from the combustion model to a two-dimensional cellular automaton model with a nearest neighbor updating rule, using lattices of size 256×256 and 1024×1024 . In order to mimic the KAMJ theory as closely as possible, we defined the CA model as follows: (1) no disorder in the front region, i.e., the simulations were done with a

uniform background ($c = 1$), (2) updates were done using strictly nearest neighbor interactions for linear growth, i.e., during a single update the “fire” (reaction front) will spread only to the “unburned” (susceptible) nearest neighbors, and the burning site itself will turn into “ashes” (inert). Simulation were done with both homogeneous and heterogeneous nucleation. The CA model matched exactly the KAMJ result for the volume fraction, (Eq. (3)) and Sekimoto’s result for the structure factor (Eq. (6)), as expected.

The constant growth velocity, and the scaling of $X(t)$ might suggest that the structure factor exhibits scaling, with the characteristic length increasing linearly in time, $L(t) \sim t$. However, as is evident from Figs. 3a and 3b, this turns out not to be the case. This can also be seen from Eq. (6). At early times the structure factor follows approximately $\sim t^5$, and for late times it falls off exponentially. The reason is that the KAMJ theory applies to uncorrelated systems. The constant-velocity growth of a single domain is essentially due to the constant driving force of an excess chemical potential. But the distribution in sizes and in space of the droplets is due to their time and position of nucleation, implied by the nucleation rate. These two time scales are not proportional to each other, so no scaling results.

V. SUMMARY

To conclude, we have studied the connection between the classical theory of nucleation and growth, and a model of slow combustion. We find that the reaction occurs with constant disorder dependent velocity with a linear scaling for the characteristic length $L(t)$ for the individual growing clusters. We have studied the structure factor of the temperature field and found good agreement with the theoretical predictions.⁽¹⁶⁾ These results could be tested in a two-dimensional reaction-diffusion cell, or simply by slowly burning uncorrelated paper, i.e., with insignificant convection.

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