Kinetic Roughening in Slow Combustion of Paper

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We present results from an experimental study on the kinetic roughening of slow combustion fronts in paper sheets. The sheets were positioned inside a combustion chamber and ignited from the top to minimize convection effects. The emerging fronts were videotaped and digitized to obtain their time-dependent heights. The data were analyzed by calculating two-point correlation functions in the saturated regime. Both the growth and roughening exponents were determined and found consistent with the Kardar-Parisi-Zhang equation, in agreement with recent theoretical work. [S0031-9007(97)03836-2]

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Kinetic roughening of driven interfaces is a ubiquitous phenomenon in nature, ranging from surface growth to the propagation of fronts in random media [1]. Extensive theoretical work of the last decade has led to a classification of these phenomena according to the asymptotic behavior of the scaling properties of such quantities as surface roughness. In many cases it has been shown that such systems can be described by local growth equations of the form

$$\frac{\partial h(\vec{r},t)}{\partial t} = \nu\nabla^2 h(\vec{r},t) + \frac{\lambda}{2}[\nabla h(\vec{r},t)]^2 + F + \eta\,, \quad (1)$$

where $h(\vec{r},t)$ is a height variable in $d+1$ dimensions, ν and λ are constants, F is the driving force, and η is a noise term. In the special case where η is white noise, Eq. (1) becomes the well-known (thermal) Kardar-Parisi-Zhang (KPZ) equation [2]. Its scaling exponents are exactly known in two dimensions. The so-called growth exponent β characterizing the early-time behavior of roughening equals $1/3$, and the roughness exponent χ that characterizes the spatial extent of roughening equals $1/2$, with $\chi + \chi/\beta = 2$. If $\lambda = 0$, Edwards-Wilkinson (EW) behavior occurs, with $\beta = 1/4$ and $\chi = 1/2$ [3]. In the case that noise depends on the height variables, Eq. (1) displays a depinning transition at some critical F_c, above which the average velocity vanishes as $v \propto (F - F_c)^\theta$. At and close to depinning, the behavior predicted by Eq. (1) depends on whether or not the nonlinear term with the prefactor λ is present. If $\lambda > 0$ at F_c, the scaling exponents at F_c can be mapped to the directed percolation depinning (DPD) model, yielding $\beta = \chi \approx 0.633$ at 2D ("quenched KPZ"), while for $\lambda = 0$, we have $\beta \approx 0.88$ and $\chi \approx 1$ ("quenched EW") [4,5]. Above F_c the quenched noise becomes asymptotically irrelevant and thermal noise limit is recovered [4].

The nature of kinetic roughening has also been under intense experimental scrutiny. Experiments on surface growth, erosion, step roughening, and related processes that should in principle be described by the KPZ equation in the appropriate regimes have failed to give conclusive evidence in favor of the thermal KPZ universality class [1]. For interfaces moving in random media, the present situation is also somewhat unclear [1]. In imbibition experiments where a liquid front is absorbed into a paper sheet, behavior consistent with DPD has been reported [6]; however, nonuniversal behavior has also been seen [7]. Perhaps the clearest evidence to date has come from fracture experiments in random media [8,9]. These have produced results ($\chi \approx 0.63$–0.72) consistent with those for a directed polymer in a random medium, a problem that can be mapped to the universality class of the 2D KPZ equation.

A promising candidate for studying kinetic roughening phenomena is a slow, flameless burning process in a random medium, which is most easily realized by slow combustion of paper sheets. Zhang *et al.* [10] have, in fact, performed such an experiment, and by analyzing the scaling of the surface width $w^2(L,t) \equiv \langle \overline{(h - \bar{h})^2} \rangle \sim L^{2\chi}$ they obtained $\chi = 0.71(5)$, a value rather close to the DPD model and much larger than that predicted by the KPZ equation. However, recent theoretical work on the problem using a continuum phase-field model of slow combustion [11,12] and simple cellular automata models of "forest fires" [5,13] demonstrate both numerically [5,11] and analytically [12] that, for the slow combustion of a uniformly random distribution of reactants, the kinetic roughening of flame fronts should be described by the KPZ equation.

We report in this Letter first results on a new experimental study of the process of slow, flameless combustion of paper. Our aim has been to create a well-controlled environment for paper burning and to study and eliminate external factors that may have influenced the results in Ref. [10]. We demonstrate that it is indeed possible to find a regime where results consistent with the KPZ universality class can be obtained in a reproducible manner.

The experimental setup shown in Fig. 1 consists of a combustion chamber, a video camera connected to a recording system, and a computer. One side of the combustion chamber is made of glass. The rest of the chamber is lined with a 50 mm layer of porous material for making the incoming air flow laminar. In the middle of the chamber there is a sample holder, designed for a maximum paper size of 600 × 400 mm. It can be rotated with respect to the adjustable air flow so that convective transfer of heat ahead of the front can be minimized. The sample holder is an open metallic frame whose sides are both lined with needles that keep the paper sheet planar during combustion. To compensate for the extra heat loss at the boundaries, the sides of the sample can be heated with filaments that follow the combustion front.

As the direction of air flow in the chamber was from bottom to top, combustion fronts were ignited from the top end of the paper by a heating wire. The emerging flame fronts were recorded with a charge coupled device (CCD) camera whose effective resolution was 752 × 582 pixels. For the typical 300-mm-wide and 500-mm-long paper samples, the pixel size was 0.28 mm which was well below the average length of fibers, i.e., ≈1.3 mm. The video signal from the CCD camera was recorded on a Super VHS recorder. The color-coded signal was converted to digital form on a video card, and analyzed on a PC using a gray scale of 256 shades. The position of the interface was determined from the maximum brightness of the front at each point.

An important issue not to be overlooked in using paper sheets in front propagation experiments is the structure of paper. First, it was recently shown [14] that, especially for low basis weight paper, there can be nontrivial power-law correlations in the areal mass density that extend up to about 15 times the fiber length, i.e., into the cm range. Second, slow combustion fronts do not easily propagate in a material made of pure cellulose fibers only. We therefore decided to use two different, easily obtainable grades of paper: ordinary copier paper with the rather high basis weight of 80 $\mathrm{g\,m^{-2}}$, and cigarette paper with a basis weight of 28 $\mathrm{g\,m^{-2}}$. The latter burns well by slow combustion, but potassium nitrate (KNO_3) was added to the copier paper to ensure uniform propagation. The concentration of KNO_3 was kept at the very low value of 0.8(2) $\mathrm{g\,m^{-2}}$.

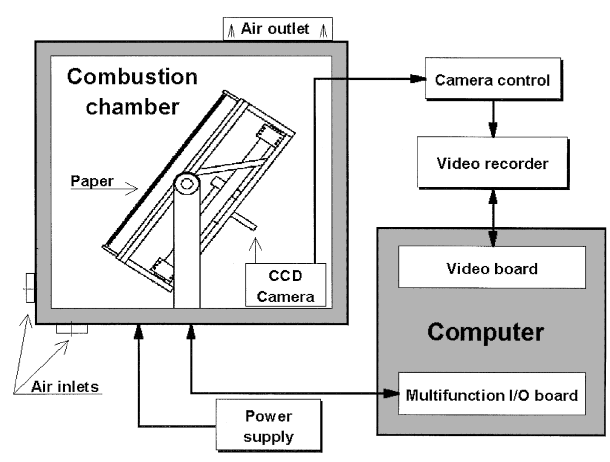

FIG. 1. A schematic diagram of the experimental setup.

In order to confirm the uncorrelated nature of the density variations in the paper samples, distributions of the calcium and potassium concentrations were both measured on the surface layer of the samples by the laser-ablation method [15]. Even though there may be concentration profiles across the thickness of the samples, there is no reason to expect qualitative changes in the density correlations in different layers [16]. The autocorrelation function of the concentration variations around the mean value was calculated from the laser ablation results and was found to collapse to the noise level within a distance of a couple of pixels.

In Fig. 2(a) we show a time series of typical digitized fronts obtained for the copier paper, while the time evolution of the surface width $w(t)$ is shown in Fig. 2(b). There is an initial transient of about 100 s or less, after which the width saturates and fluctuates around its average value of about 2 mm. After this the average velocity v of the front is constant in time to a very good approximation. The total duration of each burn was about 900 s, and in the steady-state regime $v = 0.51(5)$ mm/s.

Conceptually, the easiest quantity for analyzing the scaling behavior of the fronts is the width which scales as t^β for early times, and as L^χ in the saturated regime, where L is the system size. In the present case, the rather rapid saturation of $w(t)$ prevented us from using this quantity to estimate β. For $w(L)$ one large system is usually used, and $w(\ell)$ is calculated for subsystems of sizes $\ell \leq L$. To test how this procedure works in a system with aperiodic boundary conditions, we performed computer simulations for the restricted solid-on-solid (RSOS) growth model [17] that easily gives the KPZ exponents in $d = 1 + 1$. Even for systems as large as $\mathcal{O}(10^4)$, however, rather poor scaling of $w(\ell)$ was found.

A much better way of estimating the scaling exponents is to use the two-point correlation function

$$C(r,t) = \langle \overline{[\delta h(r_0,t_0) - \delta h(r_0 + r, t_0 + t)]^2} \rangle, \quad (2)$$

with $\delta h \equiv h - \bar{h}$ and the brackets denote an average over configurations and the bar an average over each system (sheet). Through this quantity, one can define the two functions $G(r) = C(r,0) \sim r^{2\chi}$, and $C_s(t) = C(0,t) \sim t^{2\beta}$, where in the saturated regime $G(r)$ can be averaged over all times (configurations), and $C_s(t)$

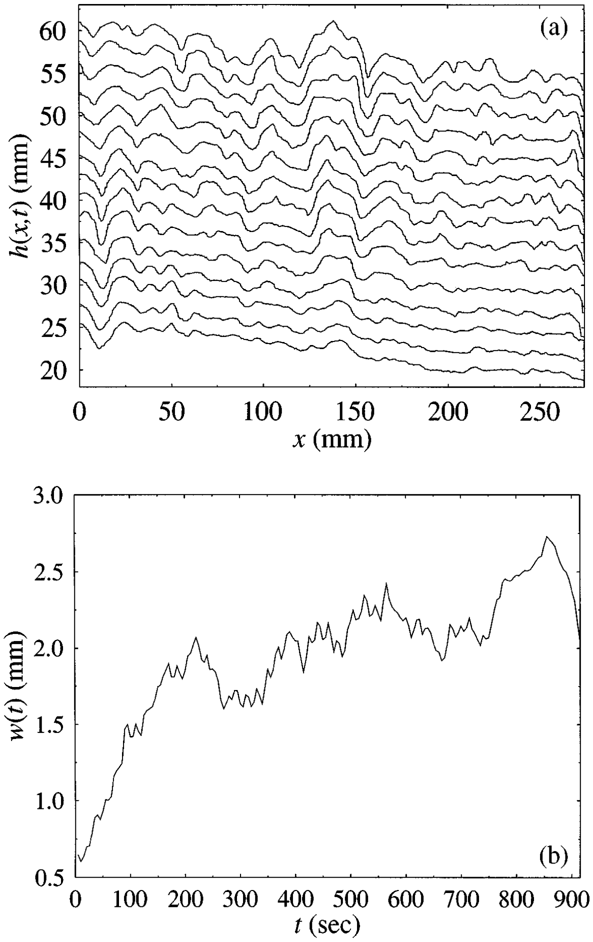

FIG. 2. (a) A series of successive digitized flame fronts taken every 5 s following the ignition of copier paper. (b) Evolution of the time-dependent surface width $w(t)$.

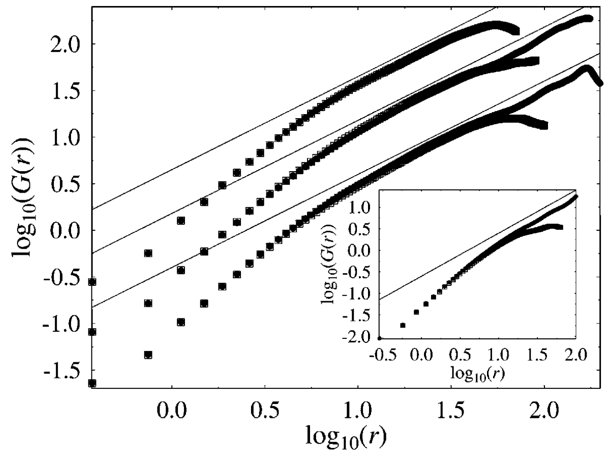

FIG. 3. The spatial correlation function $G(r)$ for three different burns of the copier paper (data have been shifted for clarity and the units are in mm). Filled circles denote the case where the average global tilt of the interface has been subtracted out. The solid lines denote $2\chi = 1$. Inset shows corresponding data for the cigarette paper.

over all spatial points. For the RSOS model, using these quantities instead of $w(\ell)$ gives very good scaling and estimates of χ and β. Most importantly, using these two functions we can obtain *independent* estimates of the two scaling exponents.

A series of typical spatially dependent correlation functions $G(r)$ for the copier paper are shown in Fig. 3. That these results are not affected by boundary effects was checked by systematically removing boundary pixels in the calculations. For short distances up to about 20 pixels or 7.5 mm, the scaling is rather poor, with an effective exponent of about 0.8. However, beyond these distances $G(r)$ scales well up to about 20 cm in the best cases, with an average exponent $\chi = 0.48(1)$ from the curves in Fig. 3. The behavior of the quantity $w(\ell)$ (not shown here) is consistent with $G(r)$. Up to $\ell \approx 1.8$ cm, the scaling is not very good ($\chi \approx 0.7$), but for larger values of ℓ, there is about 1 order of magnitude of scaling where χ is consistent with $1/2$. The finite width of the sheets prevented us from obtaining a more extended scaling regime.

In Fig. 4 we show the data for $C_s(t)$, with configurations taken every five seconds in the saturated regime. At early times up to about 50–100 s, the scaling is not very good, and one obtains an effective exponent of 0.40–0.46. However, from 100 s upwards scaling is well obeyed, and averaging over the curves in Fig. 4 gives $\beta = 0.32(1)$.

The second set of experiments on the cigarette paper gave results consistent with those for the copier paper despite the fact that the cigarette paper is strongly anisotropic and may contain nontrivial correlations. An example of the data is shown in Fig. 3 in the inset. In this case, the velocity was higher with $v = 1.64(5)$ mm/s and the width saturated in few tens of seconds following ignition. Although the overall scaling of both $G(r)$ and $C_s(t)$ is not as good as for copier paper, the scaling exponents tend towards the KPZ values asymptotically. This is a good check on the consistency of our experimental results.

The results obtained independently for the two exponents and the two grades of paper strongly support the

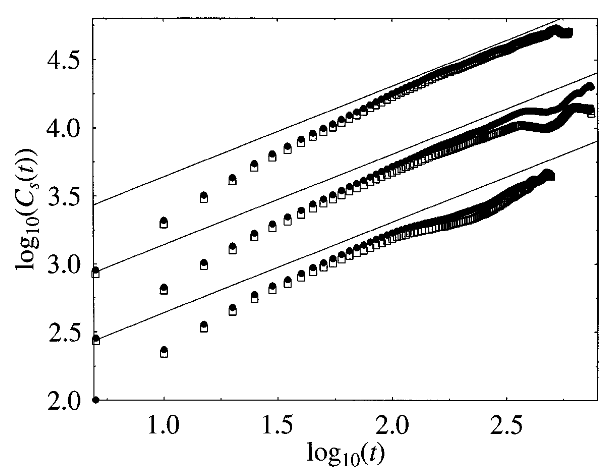

FIG. 4. Time-dependent correlation functions $C_s(t)$ for the data used in Fig. 3. The solid lines denote $2\beta = 2/3$.

conclusion that for slow combustion in uniformly random media, the kinetic roughening of the fronts is described by the KPZ equation with thermal noise. In particular, the scaling exponents obtained here definitely rule out the much larger DPD values of $\chi = \beta = 0.633$. Unfortunately, the range in which we are able to observe scaling is limited [18], and physical restrictions in the current experimental setup prevent us from significantly increasing the width of the paper sheets and thus extending the scaling regime.

As regards the earlier experiment by Zhang *et al.* [10], we have so far no definite explanation for why $\chi = 0.71(5)$ was obtained from $w(\ell)$ for all values of ℓ measured (β was not estimated). It is possible that physically different regimes exist in combustion experiments [19]. This issue warrants further investigation.

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[18] The roughness at small scales is influenced by several factors. First, nontrivial spatial correlations of areal mass density in paper up to at least the fiber length [14] increase the effective exponents at small scales. Second, the diffusion of heat in paper and in the air surrounding the front may affect short wavelength scaling. Third, finite spatial resolution in digitization also smooths out finer details at small scales. Thus, it is not surprising that scaling is well obeyed only for lengths well beyond 1 mm scales.
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