Scaling and Noise in Slow Combustion of Paper

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We present results of high resolution experiments on kinetic roughening of slow combustion fronts in paper, focusing on short length and time scales. Using three different grades of paper, we find that the combustion fronts show apparent spatial and temporal multiscaling at short scales. The scaling exponents decrease as a function of the order of the corresponding correlation functions. The noise affecting the fronts reveals short range temporal and spatial correlations, and non-Gaussian noise amplitudes. Our results imply that the overall behavior of slow combustion fronts cannot be explained by standard theories of kinetic roughening.

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Rough interfaces in nature are ubiquitous: a growing crystal surface, a droplet of coffee on a napkin, a flame front in a combustible material or a penetrating flux in a thin film superconductor all are phenomena where rough surfaces with nontrivial scaling properties may emerge. Experiments and theory on the kinetic roughening of driven surfaces have received a lot of attention. Despite considerable efforts and progress on the theoretical side, many of the experiments in kinetic roughening can be explained only partially by the present models [1–3].

In many cases of interest, the kinetic roughening of a driven interface can be described by a local equation of motion of the Kardar-Parisi-Zhang (KPZ) type [4],

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

Here $h(\vec{r},t)$ is a height variable in $d+1$ dimensions, ν and λ are constants, F is the driving force, and η is a noise term. The essential feature of Eq. (1) is the nonlinear term $\propto \lambda$, which is due to the local tilt dependence of the growth velocity of the interface. The scaling properties can be described most simply in terms of the surface width $w^2(L,t) \equiv \langle\overline{(h-\bar{h})^2}\rangle$, where the overbar and brackets denote spatial and noise averaging, respectively. For self-affine interfaces, this quantity typically scales as $t^{2\beta}$ for early times, and as $L^{2\chi}$ for late times for a finite system of size L. The quantities β and χ are called the growth and roughness exponents, respectively.

The character of the noise η has a major influence over the scaling exponents. If the noise is white, the scaling exponents of the $(1+1)$-dimensional "thermal" KPZ (TKPZ) equation are $\beta = 1/3$ and $\chi = 1/2$. If the noise depends on $h(x,t)$ Eq. (1) displays a depinning transition at some critical F_c, below which the average velocity vanishes. If $|\lambda| > 0$ at F_c, the problem can be mapped to the directed percolation depinning (DPD) model, yielding $\beta = \chi \approx 0.633$ and corresponding to "quenched" KPZ. On the other hand, for $\lambda = 0$ at F_c, $\beta \approx 0.88$ and $\chi \approx 1$ [5,6]. The moving interface just above F_c is not self-affine, has effective $\beta \approx \chi \approx 0.75$ for short length and time scales; the quenched noise becomes asymptotically irrelevant and the thermal case is recovered [5].

Various experiments on kinetic roughening have mostly failed to demonstrate TKPZ scaling. It has been suggested that this could result from nontrivial effective noise. If there are power-law correlations in the noise, it has been shown that the scaling exponents increase from the TKPZ values for some range of the correlation exponents [7]. Exceptional fluctuations in the amplitude η of the noise, such that the corresponding distribution function is $P(\eta) \sim \eta^{-(1+\mu)}, \eta \geq 1$, are another candidate. Below a critical value of $\mu_c \approx 4$, the roughness exponent depends on μ and $\chi(\mu = 3) \approx 3/4$ [8]. Such fluctuations in the noise have been demonstrated in fluid-flow experiments [9]. Computer simulations [10] have shown that the KPZ equation then displays *multiscaling* for short length scales. For $\mu = 3$, the qth order spatial correlation functions have short range (SR) roughness exponents that decrease from about 0.76 ($q = 1$) to 0.15 ($q = 9$). Beyond a crossover length, the interface is self-affine with $\chi \approx 0.76$ independent of q.

We have recently shown that asymptotic TKPZ behavior can be obtained in careful experiments on slow combustion fronts in paper [11,12], as predicted theoretically [6,13]. In an earlier experiment [14] $\chi = 0.71(5)$ was found and interpreted in terms of the moving phase of the DPD model. Very recently, a crossover to TKPZ scaling was also seen in kinetic roughening of penetrating flux fronts in high-T_c thin film superconductors [15]. The SR scaling was purported to be the same as that reported in Ref. [11], and due to a DPD phase close to depinning [16]. However, our preliminary results on the SR scaling of slow combustion fronts [12] do not support this suggestion nor the DPD predictions.

In this Letter, we present results of high-resolution spatial and temporal measurements of the kinetic roughening

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of slow combustion fronts in paper. This allows us to clarify the *short range and short time* behavior of the steady-state fronts, below the crossover scales to the asymptotic TKPZ exponents [11]. We use two different types of copier paper [11], and a lens paper similar to that in Ref. [14]. In *all three cases,* we find that, up to length scales r_c and times t_c, the fronts obey apparent spatial and temporal multiscaling with exponents incompatible with Eq. (1). For copier (lens) papers $r_c \approx 5(11)$ mm and $t_c \approx 25(2.3)$ s. The noise at the interface exhibits SR temporal and spatial correlations, and has a non-Gaussian amplitude distribution with indications of a power-law tail. The tail exponent $\mu \approx 3$ for small sampling time intervals and increases with increasing interval. The correlations in space and time persist roughly up to r_c and t_c. These results demonstrate that the SR behavior is not described by a KPZ (or a DPD) type of an equation, as suggested in Ref. [17].

The experimental setup comprises a combustion chamber combined with computer control [12]. We have considerably enhanced the resolution. The propagating front is recorded with three CCD cameras, each with an effective resolution of 768×548 pixels. The combined picture covered an area of size 300×74 mm^2. The frames were recorded using a multilevel gray scale. A single-valued front line was fitted into the recorded brightness profile. The cylindrical image distortions caused by the lenses were corrected using nonlinear warping. For a 390 mm wide and 500 mm long sample, with a sample-to-camera distance of 70 mm, the pixel size was 0.135 mm. This was an order of magnitude below the average length of the fibers.

The basis weights of the three grades of paper used were 70, 80, and 9.1 g^{-2} (lens paper) [18]. Slow combustion fronts do not easily propagate in a material of pure cellulose fibers. Copier papers contain calcium carbonate ($CaCO_3$) with a high heat capacity, which in slow combustion enhances front propagation. In addition, potassium nitrate (KNO_3) was added as an oxygen source to ensure a more uniform propagation. The concentration was kept at a low level of approximately 0.8 g^{-2}. The correlations in the absorbed mass density were found to be negligible. Most of the data presented here are for the copier papers. For the lens paper the uniformity of the KNO_3 concentration necessitates careful sample preparation.

First, we discuss the scaling of the fronts as given by the qth order two-point correlation functions

$$C_q(r,t) = \langle \overline{|\delta h(r_0,t_0) - \delta h(r_0 + r, t_0 + t)|^q} \rangle, \quad (2)$$

with $\delta h \equiv h - \bar{h}$ and the bar denotes average over an instantaneous front in the steady-state regime, and the brackets over all fronts and burns. The corresponding temporal and spatial correlation functions are shown in Figs. 1(a) and 1(b) for the copier papers. As expected [11], beyond a crossover length of $r_c = 5.2(1)$ mm, well-defined asymptotic roughness exponents $\chi = 0.49(2)$ (70 g^{-2}) and 0.44(5) (80 g^{-2}) emerge which extend beyond 100 mm and are compatible with the TKPZ value

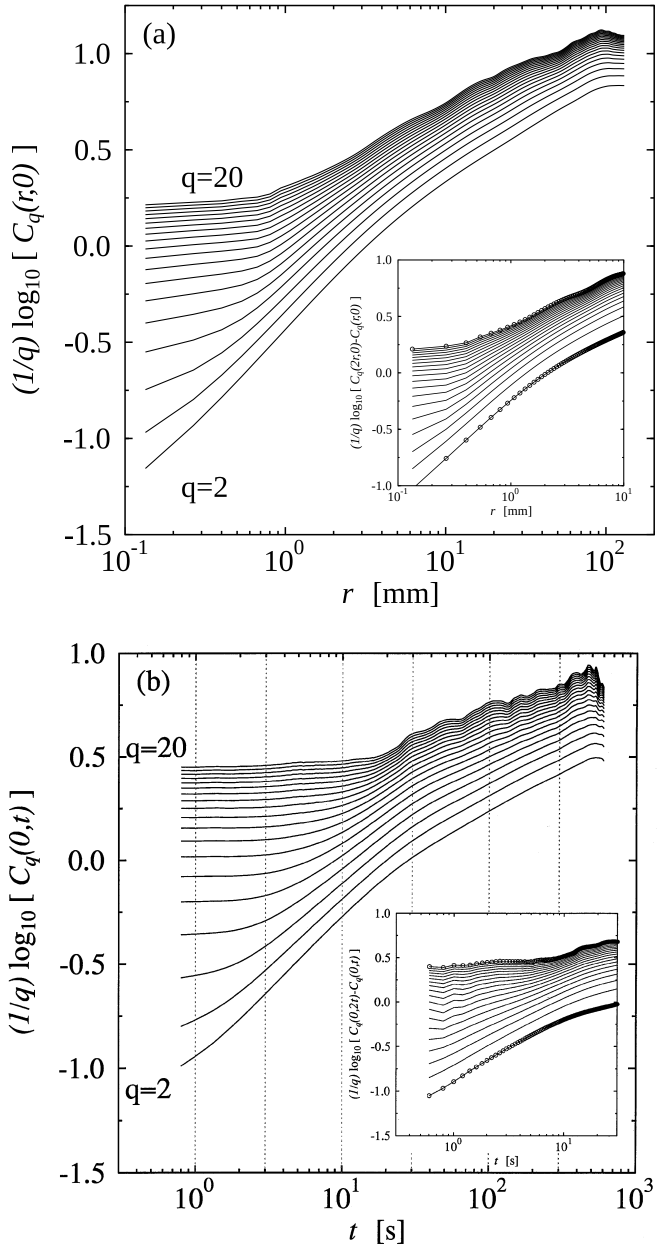

FIG. 1. (a) The qth order spatial correlation functions $C_q(r,0)$ averaged over ten burns for the 70 g^{-2} grade. Inset shows the SR behavior. Fits for the χ_{SR}'s were done up to 4 mm. (b) The qth order temporal correlation functions $C_q(0,t)$ averaged over 20 burns for the 80 g^{-2} grade. Inset shows the SR behavior. Fits to get β_{SR}'s were up to $t = 10$ s.

of $\chi = 1/2$. The interesting feature in the data is now the existence of a SR regime with roughness exponents $\chi_{SR} = \chi_{SR}(q)$. In Fig. 2 we plot these effective exponents vs q. They decrease from 0.85(6) (70 g^{-2}) and 0.87(5) (80 g^{-2}) at $q = 2$ until leveling off towards $\chi = 1/2$. Thus, there is apparent multiscaling of the fronts at SR. For the lens paper the crossover length is $r_c = 11.1(1)$ mm, and the behavior is completely analogous.

In Fig. 1(b) we show the qth order temporal correlation functions for the 80 g^{-2} grade. After $t_c = 24.6(4)$ s the TKPZ behavior is well obeyed, with $\beta = 0.35(1)$ up

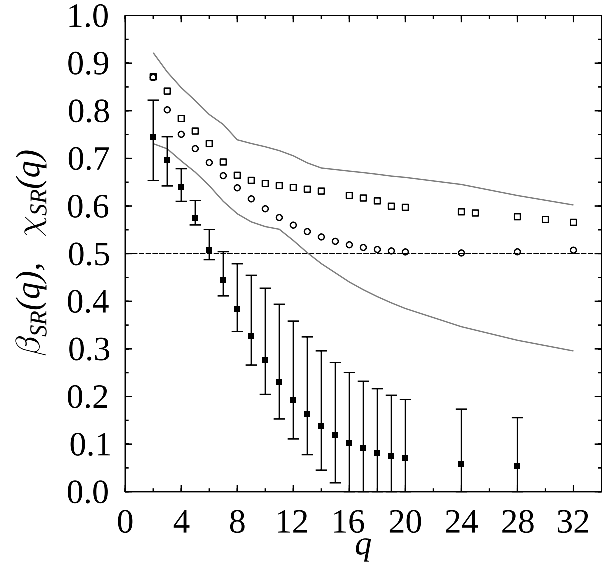

FIG. 2. The effective $\chi_{\mathrm{SR}}(q)$ vs q for the 70 g^{-2} (open circles) and the 80 g^{-2} paper (open squares). The solid lines show the maximal error bars, and the horizontal line indicates $\chi = 1/2$. The effective $\beta_{\mathrm{SR}}(q)$ vs q are shown for the 70 g^{-2} paper (filled squares).

to about 450 s. At short times the effective exponents decrease with q, from 0.75(8) at $q = 2$ to about 0.05 ($q = 28$), as shown in Fig. 2. Results for the other two paper grades are again similar.

As noise is expected to be relevant in the SR behavior, we discuss next the nature of the effective noise at the interfaces. We measured the noise amplitudes using [9] $\eta(x,t) \equiv \tilde{h}(x, t_0 + \tau) - \tilde{h}(x, t_0)$, where $\tilde{h}(x,t) = h(x,t) - \bar{h}(t)$, as a function of the time interval τ. The corresponding amplitude distributions $P(|\eta|)$ are shown in Fig. 3 for the 80 g^{-2} paper. We find $P(|\eta|)$ clearly non-Gaussian with indication of power-law behavior $P(\eta) = c\eta^{-(\mu+1)}$ for large $|\eta|$. If a power law is fitted to the tails of the distributions, which otherwise appear rather Gaussian, we find that μ changes from about 2.7 at $\tau = 1$ s to 4 at 100 s. For long time intervals, the power-law regime becomes less apparent as the crossover from Gaussian behavior for $|\eta|$ small extends further.

To check the existence of spatial and temporal correlations in the noise, we also considered the two-point correlation function of velocity fluctuations $C_u(x,t) = \langle \delta u(x_0 + x, t_0 + t)\delta u(x_0, t_0)\rangle$, with $\delta u(x,t) = u(x,t) - \bar{u}(t)$, where $u(x,t) = [h(x, t + \tau) - h(x,t)]/\tau$. This was computed for different times τ. Figure 4 shows such spatial correlation functions $C_u(x, 0)$, and the inset of the figure $C_u(0,t)$. It is evident that there are nontrivial correlations in the noise. However, they are rather insignificant beyond the crossover scales r_c and t_c, and are not of power-law type. This is in agreement with the TKPZ behavior observed asymptotically, and holds true for all three paper grades.

Multiscaling arises [10] in TKPZ from power-law noise amplitudes. Although the tails of the noise amplitude distributions are of power-law type here, our exponents are different from those of Ref. [10] on *both short and long scales*. Thus, a nontrivial noise amplitude distribution alone cannot explain our results. The SR exponents cannot be explained by noise correlations either, since they are not of power-law type (Fig. 4).

In DPD type of models there also appears [5,19] a crossover from a SR (DPD) to asymptotic TKPZ scaling.

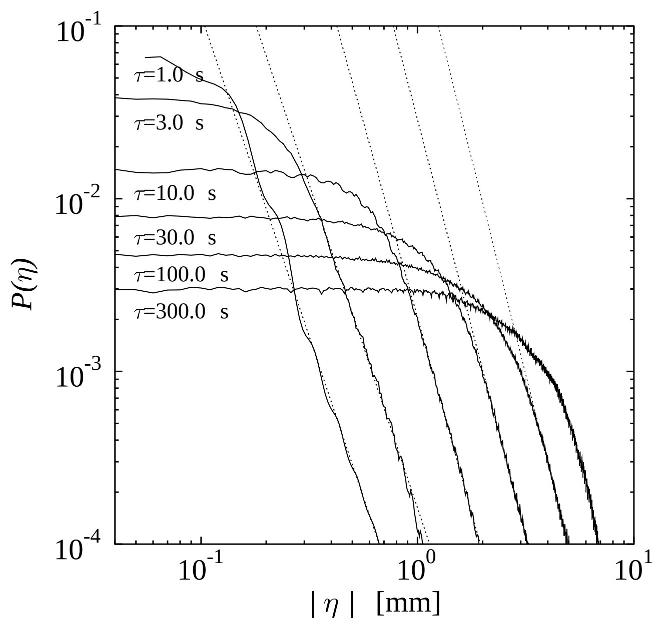

FIG. 3. A log-log plot of the distribution $P(|\eta|)$ vs $|\eta|$ with time intervals shown in the figure for the 80 g^{-2} paper. The linear fits have slopes -3.72, -3.74, -4.53, -4.81, and -5.0, left to right.

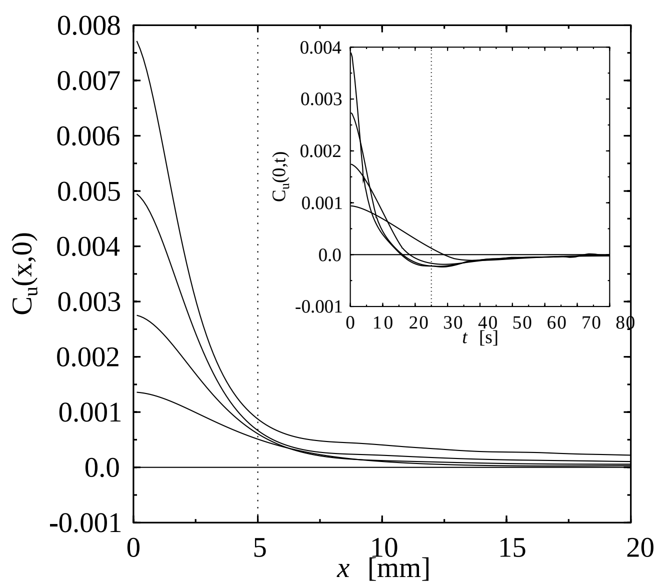

FIG. 4. The spatial velocity-fluctuation correlations $C_u(x, 0)$ for time steps $\tau = 4$, 8, 16, and 32 s for the 70 g^{-2} grade. The crossover length r_c is shown by a vertical line. The temporal correlations $C_u(0,t)$ for the 80 g^{-2} paper are shown in the inset for $\tau = 4$, 8, 16, and 32 s. The crossover time t_c is shown by a vertical line.

This can be explained [5] by Eq. (1) with quenched disorder that becomes white noise asymptotically for moving interfaces. In paper samples quenched (structural) disorder is present, but seems insignificant for the dynamics. Although the paper grades have rather different density variations, their SR exponents are very similar. Furthermore, they do not agree with the predictions for DPD type of models [20]. In thin-film superconductors DPD type of SR behavior has been observed [15]. However, it would be instructive to analyze the noise and multiscaling properties of the fronts for a consistent interpretation. Also, due to large fluctuations it is crucial to do extensive averaging, here typically over about 40 000 individual fronts. Only by such averaging, reliable estimates of the scaling exponents can be made. For the lens paper, fluctuations between different burns were particularly large, due in part to variations in the homogeneity of the KNO_3 concentration. Scaling behavior consistent with the copier papers was obtained only with homogeneous samples after sufficient averaging. This may in part explain the earlier experimental data of Ref. [14] on lens paper treated with KNO_3.

A typical mechanism leading to multiscaling in many models of kinetic roughening is the existence of avalanches in the local interface dynamics [21]. We have monitored the instantaneous velocities of the fronts and find that there are avalanches present [22]. They seem to be closely related to the formation of ash at the interface. Along the front, the burning paper separates into fragments of ash of width $\lesssim 1$ cm that curl up to strips about 2–3 cm long. Such strips fall down after about 40–60 s, with a jump in the local velocity. These scales are close to the crossover scales r_c and t_c, and are also reflected in the noise correlations of Fig. 4. The dynamical noise related to ash formation may dominate in the noise affecting the slow-combustion fronts. This would explain why quenched noise does not seem to be important.

In conclusion, our results indicate complicated scaling of slow-combustion fronts at short range. For short temporal and spatial scales we find nontrivial noise and apparent multiscaling. It seems that theoretical models of the KPZ type [Eq. (1)] cannot fully explain the slow-combustion experiments. In particular, the suggested DPD behavior [14,17] is ruled out unlike in the recent experiments on thin-film superconductors [15]. We emphasize that, to obtain reliable scaling properties for the fronts, sufficient statistics and careful sample preparation are absolutely necessary. We feel that the situation here may be analogous to that observed in the $(1 + 1)$-dimensional Kuramoto-Sivashinsky equation. In this case asymptotic TKPZ scaling is recovered only beyond crossover scales in time and space [23], whereas the microscopic physics is completely unrelated to the TKPZ behavior. The experimental indication is that it is important to consider the role of crossover scales in interpreting the role of noise, avalanches, and other possible effects in all experiments on front propagation.

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