VOLUME 92, NUMBER 25 PHYSICAL REVIEW LETTERS week ending 25 JUNE 2004

Evidence for Power-Law Dominated Noise in Vacuum Deposited CaF_2

D. R. Luhman and R. B. Hallock

Laboratory of Low Temperature Physics, Department of Physics, University of Massachusetts, Amherst, Massachusetts 01003, USA

(Received 12 February 2004; published 24 June 2004)

We have studied the surface roughness of CaF_2 vacuum deposited on glass using atomic force microscopy for film coverages spanning an order of magnitude. We find the roughness exponent $\alpha = 0.88 \pm 0.03$, the growth exponent $\beta = 0.75 \pm 0.03$, and the dynamic exponent $z = \alpha/\beta = 1.17 \pm 0.06$. Multifractality is also present, along with power-law behavior in the nearest neighbor height difference probability distribution. The results indicate noise dominated by a power-law distribution with exponent $\mu + 1 \approx 4.6$.

DOI: 10.1103/PhysRevLett.92.256102 PACS numbers: 68.35.Ct, 68.55.Jk, 81.15.Ef

Over the past several decades considerable work has been done on the kinetic roughening of surfaces. The systems studied are far from equilibrium and very often the growing interface is tortuous and well described using fractal concepts. This general field of study encompasses a diverse range of phenomena, such as thin film growth, fluid flow through porous media, the propagation of burning fronts, and the growth of bacterial colonies. Of particular interest are the critical exponents that characterize the growing rough interface [1–3].

The roughness of an interface originates through small random perturbations occurring during the growth process, or, in other words, noise. In many instances this noise was assumed to be Gaussian and uncorrelated, which led to the establishment of universality classes, each characterized by a set of critical exponents. Later work concluded that if the noise follows a power-law distribution the critical exponents are different, resulting in nonuniversal behavior [4,5]. This has been verified in $1 + 1$ dimensions using two phase fluid flow [6,7] and the propagation of slow burning fronts [8]. In this Letter, we report on experiments using CaF_2 vacuum deposited on glass that exhibit evidence of power-law distributed noise in $2 + 1$ dimensions.

Much of the current interest in this field has been stimulated by the introduction of the dynamic scaling description [9] and the stochastic growth equation due to Kardar, Parisi, and Zhang (KPZ) [10]. Starting from a flat surface of size L at time t, Family and Vicseck [9] conjectured that the interface width $w(L, t)$ has the scaling form $w(L, t) \sim L^{\alpha} f(t/L^z)$, where $w(L, t)$ is defined as

$$w(L,t) = \sqrt{\left\langle \frac{1}{L^2} \int d^2r [h(\vec{r},t) - \bar{h}(L,t)]^2 \right\rangle}. \tag{1}$$

The brackets denote an average over different samples, and $h(\vec{r}, t)$ is the height of the interface at position $\vec{r}$. The mean height of the interface is given by $\bar{h}(L, t)$. For surfaces that saturate for $t \gg L^z$, the scaling function behaves as $f(x) \rightarrow$ constant when $x \rightarrow \infty$; otherwise, $f(x) \sim x^{\beta}$. For early times, the width is related to time through

$$w(L,t) \sim t^{\beta} \qquad 1 \ll t \ll L^z \tag{2}$$

and when $t \gg L^z$, $w(L, t) \sim L^{\alpha}$. The exponents are not independent, rather they are related through $\beta = \alpha/z$. The set of α, β, and z constitute the critical exponents and are designated the roughness exponent, the growth exponent, and the dynamic exponent, respectively [1].

The KPZ equation [10]

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h + \frac{\lambda}{2}(\nabla h)^2 + \eta \tag{3}$$

is a nonlinear equation proposed to describe interface growth in the presence of noise. Here ν is an effective surface tension and λ describes the strength of lateral growth. The noise is introduced through η, which is usually assumed to be Gaussian with $\langle \eta(\vec{r}, t)\rangle = 0$ and uncorrelated satisfying $\langle \eta(\vec{r}, t)\eta(\vec{r}\,', t')\rangle \sim \delta(\vec{r} - \vec{r}\,')\delta(t - t')$. The critical exponents of Eq. (3) can be found analytically in $1 + 1$ dimensions ($\alpha = 1/2, \beta = 1/3$) and estimated numerically in $2 + 1$ dimensions ($\alpha = 0.38$, $\beta = 0.24$) [1]. An additional restriction due to Galilean invariance is placed on the exponents in all dimensions resulting from Eq. (3) [11–13],

$$\alpha + \frac{\alpha}{\beta} = 2. \tag{4}$$

Many experiments aimed at measuring the scaling exponents of a system found $\alpha > 1/2$. This led to the proposal of Zhang [4,5], who realized that a system dominated by noise that follows a power-law distribution, rather than a Gaussian, results in different exponents. In a $D + 1$ dimensional system with an uncorrelated power-law probability distribution of the form

$$P(\eta) \sim \begin{cases} \eta^{-(\mu+1)} & \text{for } \eta > 1 \\ 0 & \text{otherwise} \end{cases} \tag{5}$$

the scaling exponents of the KPZ equation were estimated to be [5,14]

$$\alpha = \frac{D+2}{\mu+1}, \qquad \beta = \frac{D+2}{2\mu - D}. \tag{7}$$

Later work provided theoretical evidence that these equations were in fact exact [15]. These equations are believed to be valid for $D + 1 < \mu < \mu_c$, where there is a crossover to Gaussian-type behavior at μ_c. In 2 + 1 dimensions, it appears that only one numerical simulation has confirmed the result for β in Eq. (7) [16], and no experimental evidence has been put forth for the existence of power-law distributed noise.

The samples we study consist of CaF_2 vapor deposited at 0.2 nm/sec onto glass substrates. A sequence of samples of film coverages, d, were prepared and are identified by $30 \leq d \leq 520$ nm, constituting two sets of samples. The value of the film coverage, d, used to identify each sample was determined from the deposition observed on a quartz crystal microbalance, measured simultaneously with the deposition on each glass substrate. The coverage, d, is reported here as a film thickness presuming the bulk density of CaF_2. Profilometer measurements of the actual thickness of the CaF_2 films, d_p, were made on each of the studied samples and confirm that the deposited CaF_2 is porous, with a constant porosity $\phi = 0.462 \pm 0.006$, independent of coverage. The two quantities d and d_p are related by $d_p = d/(1 - \phi)$. All depositions were done at a constant rate, hence $t \sim d$. Set I samples were fabricated solely for surface analysis, while set II samples were used for low temperature investigations [17], as well as surface analysis. The two sets overlapped in thickness range, providing multiple samples for certain thicknesses. The set II samples had narrow Ag and Al bands deposited on the glass surface prior to the deposition of CaF_2. The thin films of metal constituted a very small percentage of the surface area of the sample. All samples were stored under vacuum after fabrication.

Images (512 pixels × 512 pixels) of the surfaces were obtained using tapping mode atomic force microscopy (AFM) under ambient conditions. Ensemble averages were taken over two images of each surface. Images were captured at different locations, both of which were far away from the CaF_2 film edges and the metal deposits in the case of the samples in set II. The size of the images taken using AFM varied due to the change in the size of the surface structures as d changed and also were varied to facilitate comparison. The scan size of the set II samples was $2\ \mu m \times 2\ \mu m$ and the scan size for the set I samples was $2\ \mu m \times 2\ \mu m$ for $d \geq 90$ nm and $1\ \mu m \times 1\ \mu m$ for $d \leq 125$ nm. The full experimental details will be presented elsewhere [18].

As is customary, we define the qth order moment of the surface as[1]

$$C_q(\ell) = \left\langle \left\langle \frac{1}{L^2} \int d^2 r |h(\vec{r}, t) - h(\vec{r} + \vec{\ell}, t)|^q \right\rangle \right\rangle_{|\vec{\ell}|=\ell}, \quad (7)$$

where the inner brackets denote an ensemble average and the outer brackets denote a radial average. $\vec{\ell}$ is a vector that determines the distance between two points on the surface. For ℓ less than the correlation length, the moments scale as $C_q \sim \ell^{qH_q}$, where H_q is a function of q and $H_2 = \alpha$ and $C_2(\infty) \sim t^{2\beta}$ as $\ell \rightarrow \infty$ [1]. If H_q is constant, then the surface is self-affine and $H_q = \alpha$; if H_q changes with q, then the surface is multifractal. Numerical work [19] has suggested that a system dominated by power-law noise leads to multifractality.

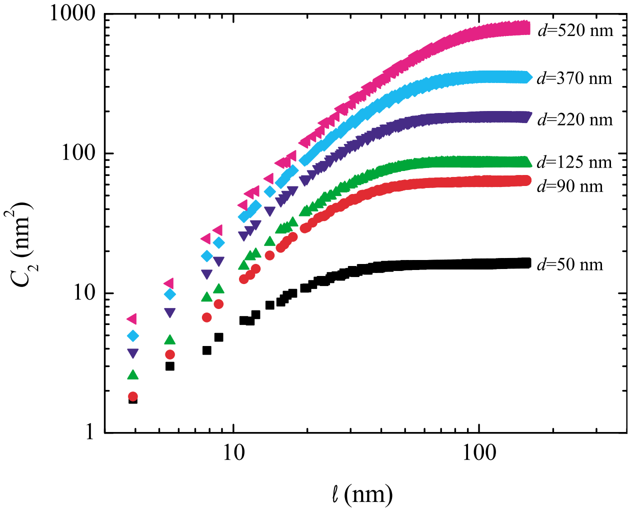

FIG. 1 (color online). C_2 versus ℓ for several of the $2\ \mu m \times 2\ \mu m$ sets of images.

Figure 1 shows values of $C_2(\ell)$ versus ℓ for a representative set of samples. 2α is defined to be the slope of a linear fit in the region of small ℓ. All of the values of α obtained from these experiments are shown in Fig. 2. α is relatively constant, except for the values from two samples (set II, $d = 50$ nm and 520 nm) and the average

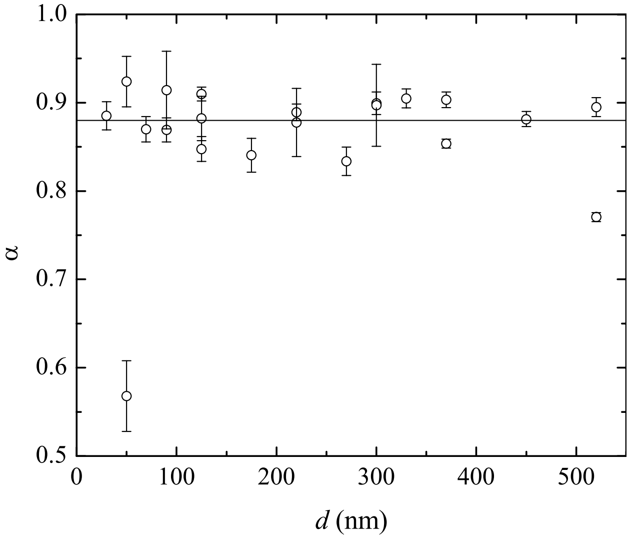

FIG. 2. α versus d for all surfaces. The solid line indicates the average value, $\bar{\alpha} = 0.88 \pm 0.03$, excluding the two anomalously low values.

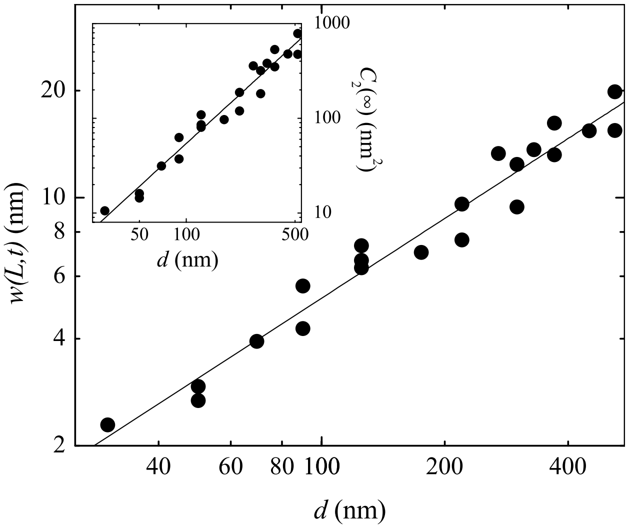

FIG. 3. Plot of $w(L, t)$ versus d (i.e., time) on a log-log scale for all sets of images. The inset shows $C_2(\infty)$ versus d. The solid line in both plots is a functional fit to a power law resulting in the identical result $\beta = 0.75 \pm 0.03$.

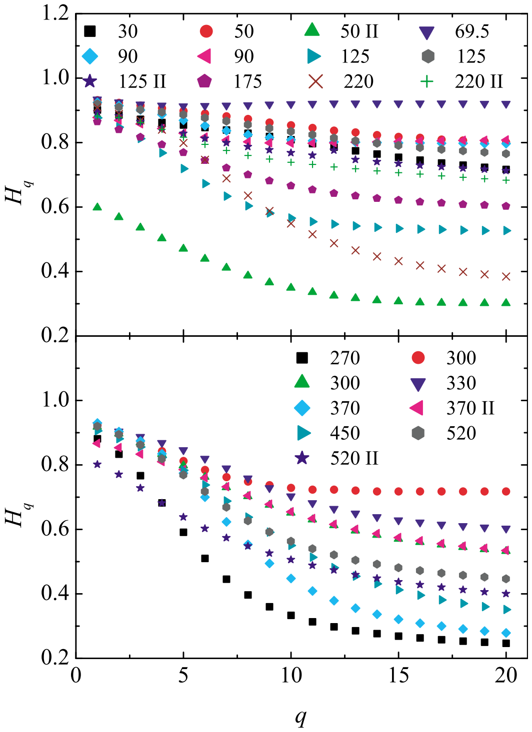

FIG. 4 (color online). H_q versus q for all samples. The data are shown in two panels for clarity. The numbers in the legend refer to the thickness of the film, d, in nanometers. The label II refers to set II, and those without this designation are from set I.

of all values of α, excluding the two anomalous points, as shown by the solid line in the figure is $\bar{\alpha} = 0.88 \pm 0.03$. The interface width of each sample, calculated using Eq. (1), is shown in Fig. 3, where the solid line is a linear fit to the data whose slope gives $\beta = 0.75 \pm 0.03$ [See Eq. (2)]. The inset of Fig. 3 shows $C_2(\infty)$ versus d, where $C_2(\infty)$ is taken from the saturation values of $C_2(\ell)$. The line is a fit to the data yielding an identical value of $\beta = 0.75 \pm 0.03$. Evidence for multifractal behavior is clearly present in Fig. 4 from the fact that for nearly all the data sets there is a decrease in H_q in the region $q =$ 2.5–5.0, after which H_q approaches a constant value for each set of images. Again, the set II sample with $d = 50$ nm is atypical, as is the set II sample with $d = 520$ nm but to a lesser extent.

If our system exhibits power-law noise, then solving Eq. (7) with our measured values of α and β should produce a consistent value for μ. Using $\bar{\alpha} = 0.88 \pm 0.03$ yields $\mu = 3.5 \pm 0.2$, while using $\beta = 0.75 \pm 0.03$ yields $\mu = 3.7 \pm 0.1$; both values of μ are in statistical agreement. Barabási *et al.* [19] have argued that noise that follows a power-law distribution leads to a phase transition in the H_q spectrum at $q = \mu$. The transition shown [19] in H_q is a downward step starting at $q = \mu$, similar to those seen in Fig. 4, although our steps are smaller and broader. We can use the downward turn in our H_q spectra to estimate μ. The transition points are not well defined; accordingly, we estimate $\mu = 2.5$–5.0, which is in agreement with the above values. We also point out that our results satisfy Eq. (4), with $\alpha + \alpha/\beta = 2.05 \pm 0.07$.

Lam and Sander [20] have shown that for systems dominated by power-law noise the probability distribution for nearest neighbor height differences, δ, is given by $P(\delta) \sim \delta^{-\mu}$ for large δ. We define δ for our images as

$$\delta = \frac{|h(\vec{r}, t) - h(\vec{r} + \vec{r}_{nn}, t)|}{|\vec{r}_{nn}|}, \tag{9}$$

where $\vec{r}_{nn}$ is the vector pointing from $\vec{r}$ to the nearest neighbor. In our case, $|\vec{r}_{nn}| = 1$ pixel. Figure 5 shows $\log[P(\delta)]$ (ensemble averaged) for several typical substrates. For large d there are two regions of linearity. The linear region nearest to $\log[\delta] = 0$ is most prominent for large values of d. This region shrinks as d gets smaller and is absent for $d < 175$ nm. The values of the slope in this region are slightly scattered within the range 2.5–5, which encompasses the previously determined values of μ. The second region of linearity persists to the smallest d, with a slope ranging from -10 to -5 for $d = 30$ nm to $d = 520$ nm. This region is most likely due to an experimental upper limit in the height fluctuations.

The acceptance of the concept of power-law distributed noise has been hindered due to the lack of physical justification for its existence [2,4], although it has been shown that power-law noise can arise in models having quenched disorder [21], such as that found in the two phase fluid experiments [6,7]. Collectively, our results suggest the existence of power-law distributed noise, which dominates our system. We are not able to define and measure a noise spectrum directly as was done in several $1 + 1$ dimensional experiments [6,8]. This is

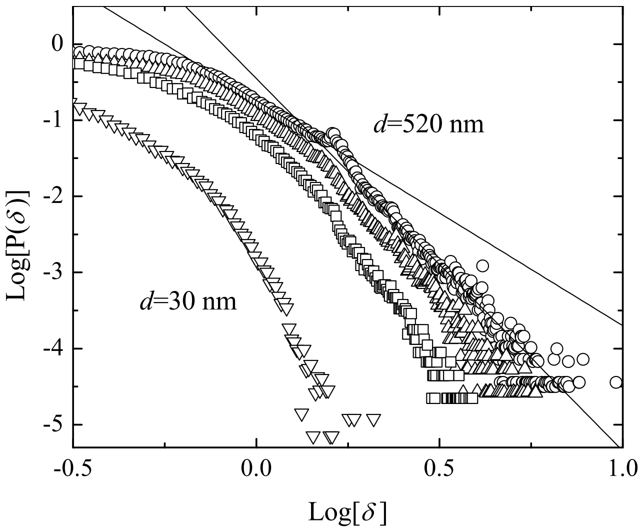

FIG. 5. Log[$P(\delta)$] versus log[δ] for $d = 30$, 175, 300, and 520 nm, corresponding to the inverted triangles, squares, triangles, and circles, respectively. There are two visible linear regions for $d \geq 175$ nm and only one for $d < 175$ nm. The solid lines show a linear fit to the each linear region for $d = 520$ nm.

because we do not measure a single interface as it grows, but rather grow an interface for a time, t, and then repeat the process for a different substrate and a different t. The result is our sets of samples, which are many realizations of the noise spectrum. The physical source of the noise in our experiments is not obvious. The interfaces do not evolve in the presence of quenched disorder; therefore the mechanism of the noise is different than in the two phase fluid experiments.

Theoretical work using a Huygens principle construction to model the columnar growth of an amorphous film has indicated that the initial conditions of a sputter deposited interface can noticeably alter the morphology of the growing surface [22]. An AFM image of a glass substrate, similar to those used here, reveals very small surface structure with $w(L, 0) = 0.4$ nm. We did, however, see two regions of power-law scaling in C_q for glass with a crossover ranging from $\ell_* \approx 20$ nm in C_1 to $\ell_* \approx 30$ nm for C_{20}, with $\alpha = 0.51 \pm 0.03$ and $\alpha = 0.19 \pm 0.03$ for $\ell < \ell_*$ and $\ell > \ell_*$, respectively. The H_q spectra of both linear regions exhibited a gentle step near $q = \mu$, as did those in Fig. 4. The step in H_q for $\ell > \ell_*$ was a decreasing function of q and its magnitude ~ 0.1, while the step for $\ell < \ell_*$ was an increasing function of q with a larger step size of ~ 0.35. The steps occur at a q which coincides with our earlier determined values of μ, but the roughness exponents are considerably different. The different values of α suggest that roughness of the CaF_2 is not a direct extension of the glass roughness. Therefore, it is not clear to what extent, if any, the initial condition of the glass played in the growth of the surfaces.

In summary, we have provided evidence for a power-law distribution of noise in vacuum deposited CaF_2 on glass. Since we were unable to measure the noise spectrum directly, we cannot check for the existence of spatiotemporal correlations, which may result in comparatively high exponents such as ours. Regardless, the evidence presented here allows the conclusion that our system is well described by the KPZ equation with noise dominated by a power-law noise distribution.

We thank K. Thompson for assistance with sample fabrication and data collection. This work was supported by the National Science Foundation under Grants No. DMR-0138009 and No. DMR-0213695 (MRSEC) and also by research funds administered by the University of Massachusetts Amherst.

[1] A.-L. Barabási and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge Unversity Press, New York, 1995).
[2] T. Halpin-Healy and Y.-C. Zhang, Phys. Rep. **254**, 215 (1995).
[3] M. Marsili, A. Maritan, F. Toigo, and J. R. Banavar, Rev. Mod. Phys. **68**, 963 (1996).
[4] Y.-C. Zhang, J. Phys. (Paris) **51**, 2129 (1990).
[5] Y.-C. Zhang, Physica (Amsterdam) **170A**, 1 (1990).
[6] V. K. Horáth, F. Family, and T. Vicseck, Phys. Rev. Lett. **67**, 3207 (1991).
[7] V. K. Horáth, F. Family, and T. Vicseck, J. Phys. A **24**, L25 (1991).
[8] M. Myllys, J. Maunuksela, M. Alava, T. Ala-Nissila, and J. Timonen, Phys. Rev. Lett. **84**, 1946 (2001); M. Myllys, J. Maunuksela, M. Alava, T. Ala-Nissila, J. Merikoski, and J. Timonen, Phys. Rev. E **64**, 036101 (2001).
[9] F. Family and T. Vicsek, J. Phys. A **18**, L75 (1985).
[10] M. Kardar, G. Parisi, and Y.-C. Zhang, Phys. Rev. Lett. **56**, 889 (1986).
[11] P. Meakin, P. Ramanlal, L. M. Sander, and R. C. Ball, Phys. Rev. A **34**, 5091 (1986).
[12] J. Krug, Phys. Rev. A **36**, 5465 (1987).
[13] E. Medina, T. Hwa, M. Kardar, and Y.-C Zhang, Phys. Rev. A **39**, 3053 (1989).
[14] J. Krug, J. Phys. I (France) **1**, 9 (1990).
[15] C.-H. Lam and L. M. Sander, Phys. Rev. Lett. **69**, 3338 (1992); C.-H. Lam and L. M. Sander, Phys. Rev. E **48**, 979 (1993).
[16] R. Bourbonnais, J. Kertèsz, and D. E. Wolf, J. Phys. II (France) **1**, 493 (1991).
[17] D. R. Luhman and R. B. Hallock (to be published).
[18] D. R. Luhman and R. B. Hallock (to be published).
[19] A.-L. Barabási, R. Bourbonnais, M. Jensen, J. Kertèsz, T. Viscsek, and Y.-C Zhang, Phys. Rev. A **45**, 6951 (1992).
[20] C.-H. Lam and L. M. Sander, J. Phys. A **25**, L135 (1992).
[21] Z. Csahók, K. Honda, E. Somfai, M. Viscek, and T. Vicsek, Physica (Amsterdam) **200A**, 136 (1993).
[22] C. Tang, S. Alexander, and R. Bruinsma, Phys. Rev. Lett. **64**, 772 (1990).