

Interface Roughening with a Time-Varying External Driving Force.

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Abstract. – We present a theoretical and numerical investigation of the effect of a time-varying external driving force on interface growth. First, we derive a relation between the roughening exponents which comes from a generalized Galilean invariance, showing how the critical dimension of the model is tunable with the external field. We further conjecture results for the exponents in two dimensions, and find consistency with data obtained through simulations of two models we expect to be in the same universality class. Finally, we discuss how our results can be investigated experimentally.

Open systems pose among the most challenging problems in condensed-matter physics. The transient dynamics and rich steady-state behaviour of systems ranging from dendrites to convective fluid cells provide a unique testing ground for the ideas and methods of nonequilibrium physics. Many such problems involve pattern formation where interfaces are present. A particularly well-defined problem of this kind is the roughening of a growing interface [1]. A great deal of theoretical progress has been made on this problem by analytic and numerical methods, following the now classic work of Kardar, Parisi, and Zhang (KPZ) [2]. They proposed a nonlinear differential equation to model the height h of a growing interface driven by an external flux of particles:

$$\frac{\partial h}{\partial t} = \nu \frac{\partial^2 h}{\partial \mathbf{x}^2} + \frac{\lambda}{2} \left(\frac{\partial h}{\partial \mathbf{x}} \right)^2 + \eta, \quad (1)$$

where ν and λ are constants, and η is a random noise which is assumed to satisfy Gaussian statistics with $\langle \eta(\mathbf{x}, t) \eta(\mathbf{x}', t') \rangle = 2D \delta^{d-1}(\mathbf{x} - \mathbf{x}') \delta(t - t')$, where D is a constant, and

the brackets denote a statistical average. The vector \mathbf{x} determines positions in a $(d-1)$ -dimensional plane⁽¹⁾ of a full space $\mathbf{r} = (\mathbf{x}, y)$, while t is the time. The term proportional to λ describes lateral growth of the interface, and plays an important role in roughening. This nonlinearity also implies that the model has no free energy.

Salient features of this system are the average velocity $\partial\langle h \rangle / \partial t \sim \text{const}$, and the interface width $W \equiv \sqrt{\overline{h^2} - (\overline{h})^2}$, where the bar denotes spatial average. The width obeys

$$W(L, t) \sim L^\chi f(tL^{-z}), \quad (2)$$

for late times and large length scales, where L is the linear size of the growing substrate, and f is a scaling function. In any dimension, eq. (1) presents an invariance, commonly referred to as «Galilean», providing an exact relationship between χ and z . In addition, for $d = 2$, a fluctuation-dissipation theorem allows one to calculate the interface exponents, $\chi = 1/2$ and $z = 3/2$ [3]. These are consistent with numerical simulations and differ from the trivial exponents obtained for $\lambda = 0$: $\chi = (3-d)/2$ and $z = 2$ [4]. A renormalization group analysis shows that the critical dimension of the model is $d_c = 3$, but that it involves an infra-red unstable fixed point. Thus d_c is thought to be a lower critical dimension for a transient which occurs in $d > 3$ between a strong-coupling and a weak-coupling behaviour.

Many justifications of this equation have been made in different contexts, particularly for sputtered growth of epitaxial layers. Here we focus on the interface of a stable phase growing at the expense of a metastable phase [5]. One example of such a system is the full field model of critical dynamics, called model A [6]. There a nonconserved order parameter field $\psi(\mathbf{r}, t)$ is the only dynamical mode. In equilibrium, at low enough temperatures, a stable interface exists between coexisting uniform phases $\psi \approx \pm 1$. If a system is prepared in this fashion and a constant external field H which is conjugate to ψ is applied, one of the two phases becomes metastable, and the interface advances through it. It can be shown that the dynamics of the interface separating those phases is described by the KPZ equation, with $\lambda \propto H$ ⁽²⁾. Systems such as the surfaces of growing dendrites also correspond to the KPZ equation, if one takes the appropriate limit of model C, where, in addition to ψ , there is a conserved field coupled asymmetrically to the order parameter. In this case, the diffusion length of the conserved field must be much larger than the system size.

The purpose of this letter is to study the interesting possibility of a *time-varying* external field $H(t)$ in model A. The mapping giving the KPZ equation still follows, but now with

$$\lambda(t) \propto H(t). \quad (3)$$

Since H is an external field its time dependence can be prescribed in any fashion. Most importantly, we expect it to be experimentally tunable, as we shall discuss below.

If one assumes now a time-dependent $\lambda(t)$ in the KPZ equation (1), there are a number of interesting properties, three of which we now describe. Our main working hypothesis will be that the scaling relation (2) still remains valid.

⁽¹⁾ Here, d is the dimension of the full space. A common notation is to write $d = (d-1) + 1$. In our notation $1+1=2$, $2+1=3$, and so on.

⁽²⁾ The mapping follows on large time scales which are, nevertheless, sufficiently small that nucleation does not take place in the metastable state.

i) The equation has an invariance under a generalized «Galilean» transformation

$$\mathbf{x} \rightarrow \mathbf{x} - \boldsymbol{\varepsilon} \int_{\text{const}}^t \lambda(t') dt' \quad \text{and} \quad h \rightarrow h + \boldsymbol{\varepsilon} \cdot \mathbf{x}, \quad (4)$$

where $\boldsymbol{\varepsilon}$ is a constant vector of infinitesimal amplitude. This implies that $\lambda(t)$ is not renormalized under the application of the renormalization group, since it is directly involved in an exact invariance of the equation. Here we consider a power law dependence of $\lambda(t)$ on time as

$$\lambda(t) = \frac{\lambda_0}{t^\alpha}, \quad (5)$$

where λ_0 is a constant, and report results for $\alpha > 0$ (we have also studied $\alpha < 0$). The invariance above implies the novel relations

$$\chi + z = 2 + z\alpha \quad \text{and} \quad d_c = 3 - 4\alpha, \quad (6)$$

where the *tunable critical dimension* is obtained from the trivial $\lambda = 0$ exponents. If the scaling hypothesis is valid, these are exact results. We note that in the asymptotic regime, $\partial\langle h(t)\rangle/\partial t \sim t^{-\alpha}$. This model has been studied previously by Lipowsky [7]⁽³⁾ in a different context.

ii) The importance of the result (6) lies in that a nonzero α could be sufficient to move to lower dimensions the phase transition which the usual KPZ equation exhibits only for $d > 3$. This will make it experimentally accessible. For such a transition, at small λ_0 we should have the trivial exponents, which satisfy $(2 - \chi)/z = \min((d + 1)/4, 1)$, while beyond some larger value of λ_0 , (6) should hold, implying $(2 - \chi)/z = 1 - \alpha$. In principle, it should also be possible to investigate this transition analytically, but we have not been able to do so. We intend to numerically study the possibility of this transition in the future.

iii) The nontrivial requirement of Galilean invariance implies that exponents are tunable in the presence of a time-varying field. In $d = 2$, a fluctuation-dissipation relation is satisfied both for $\lambda = 0$ and $\lambda = \text{const}$ (*i.e.* for $\alpha = +\infty$ and $\alpha = 0$) giving $\chi = 1/2$. We therefore conjecture that the value of χ remains $1/2$ for any intermediate $\lambda \propto 1/t^\alpha$, with $\alpha > 0$. This hypothesis and eq. (6) fix the values of the scaling exponents:

$$\chi = \frac{1}{2} \quad \text{and} \quad z = \frac{3}{2(1 - \alpha)} \quad (7)$$

for $d = 2$, with $0 \leq \alpha \leq 1/4$. For $\alpha > 1/4$ one is above the critical dimension and we expect the ideal interface results with $z = 2$, as discussed further below.

The question of a phase transition merits further discussion. For $\alpha = 0$, $d_c = 3$ is a lower critical dimension above which an infra-red unstable fixed point exists. As α is increased, the nonlinearity should become less relevant and it is likely that beyond a value α_{ucd} the tunable critical dimension becomes an upper critical dimension involving an infrared stable fixed point. Thus, there would be a phase transition above $d_c(\alpha)$ only for $\alpha < \alpha_{ucd}$. We now give an argument for the value of α_{ucd} . The difference $\chi(\alpha) - (3 - d)/2$ gives the amount the steady-state-driven roughness exceeds the ideal interface value. If the critical dimension has

⁽³⁾ This model was studied in ref. [7] to investigate wetting of a substrate. That involves particular values for the exponent α , for which λ turns out to be an irrelevant variable.

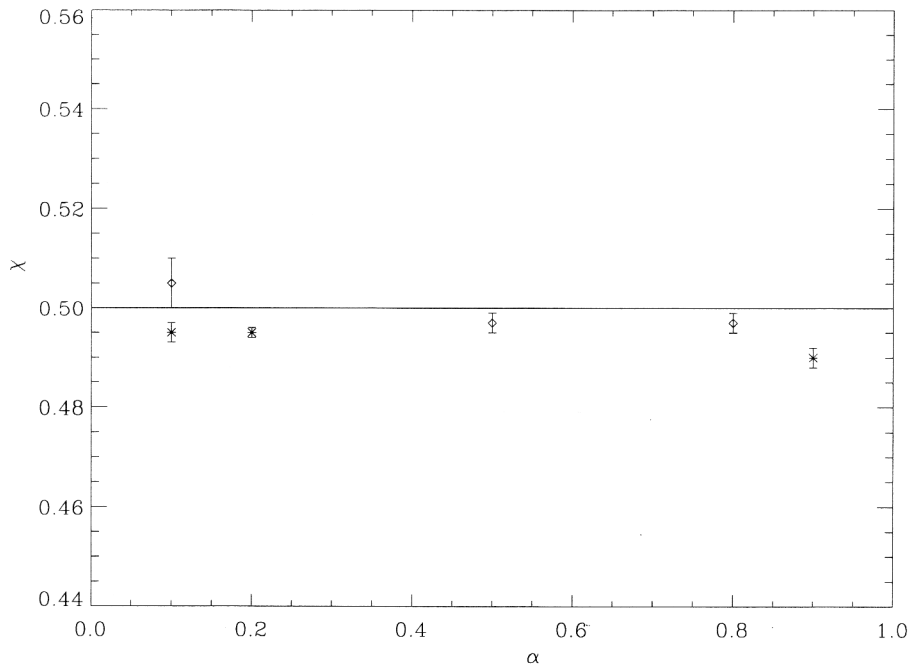


Fig. 1. – The exponent χ as a function of α from simulations of the KPZ equation (diamonds) and the GRSOS model (asterisks). KPZ data come from averages over 100 realizations of systems of sizes $L = 50, 100, 200,$ and 400 . GRSOS data have been averaged over $10^7 \div 10^8$ Monte Carlo steps for $L = 100, 200, 500$ and 1000 . The error bars are purely statistical; no systematic finite-size errors have been included.

$\chi(\alpha) - (3 - d_c)/2 > 0$, the nonlinearity is important, and we expect d_c to be a lower critical dimension, while the marginal case defines α_{ucd} by $\chi(\alpha_{ucd}) - (3 - d_c)/2 = 0$. Now, from our hypothesis equation (7) and the expression for d_c we find $\alpha_{ucd} = 1/4$ in $d = 2$, implying no phase transition because for $\alpha < \alpha_{ucd}$, $d = 2$ is below d_c . This argument rests on two conjectures, but indeed we have found no evidence for a phase transition in $d = 2$ for $\alpha > 1/4$; however, the possibility of a transition in $d = 3$ remains and will be studied in future work.

We have checked our hypothesis equation (7) numerically with two models for $d = 2$. First, we have directly integrated the KPZ equation with a time-dependent λ . Second, we have simulated the restricted solid-on-solid growth (GRSOS) model [8] using a time-varying growth rate.

The details of our numerical work are as follows. The KPZ equation, discretized as in [5], was integrated by the Euler method. System sizes ranging from 50 to 50 000 lattice sites were studied. The time step was generally taken as 0.02, and some smaller values were considered to check accuracy. Typical values for the rest of the parameters were $\nu = 3.5$, $\lambda_0 = 50$ and $D = 0.01$, and several values of α were considered. For the GRSOS model, systems of sizes up to $L = 50\,000$ were simulated too. The algebraic decay of λ was achieved by incorporating time-dependent condensation *and* evaporation rates, whose difference is proportional to λ . For both models, the known limits for $\alpha = 0$ and $\alpha = \infty$ were checked.

Direct determinations of χ were obtained by running systems of several sizes L until saturation, and then using the scaling of $W(L, t = \infty)$ with L . For both models, we obtained

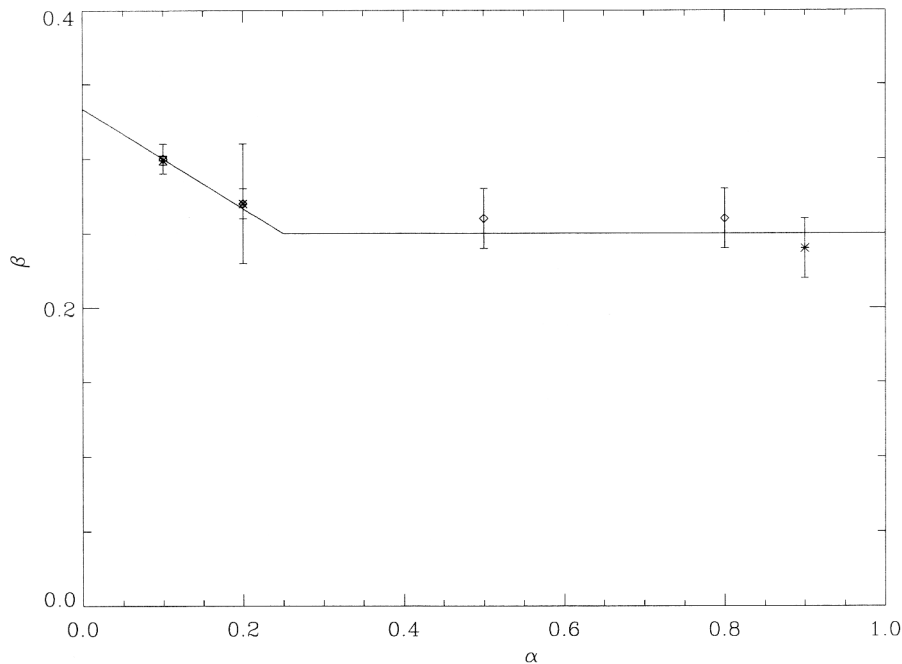


Fig. 2. – The exponent β as a function of α . Meaning of the symbols as in fig. 1. The solid line is the prediction of eq. (8). Data come from simulations of systems of size $L = 50\,000$ averaged over 30 (KPZ) and $30 \div 60$ (GRSOS) independent runs.

$\chi \approx 0.5$ for all values of α studied, as shown in fig. 1. We note that due to finite-size effects, the χ values obtained tend to be slightly below 0.5; however, this also occurs for $\alpha = 0$. Thus our results support the conjecture that $\chi = 1/2$. We have furthermore tested the scaling relations from the exponent $\beta = \chi/z$, where $W \sim t^\beta$ if $t \ll L^z$. For $d = 2$ we expect

$$\beta = \frac{1 - \alpha}{3}, \quad (8)$$

for $0 \leq \alpha \leq 1/4$ and $\beta = 1/4$ for $\alpha \geq 1/4$. To obtain β , we performed extensive calculations of the time-dependence of the width. Our results are summarized in fig. 2. It is interesting to note that it becomes increasingly difficult to extract β approaching $\alpha = 1/4$, presumably due to the vicinity of a fixed point, as discussed following item iii). However, our results for various values of α are in complete agreement with eq. (8). We also find no evidence of a possible phase transition beyond $\alpha = 1/4$, although we have not done an exhaustive search as a function of the amplitude of $\lambda(t)$.

A somewhat different way of checking our prediction for the exponents can be obtained from the *crossover scaling form* of $W(L = \infty, t)$ proposed in [5], which for $d = 2$ reads

$$W(L = \infty, t) \sim t^{1/4} g(t\lambda^\phi), \quad (9)$$

with $g(x) \sim x^{1/12}$ for large x . Theoretical arguments and computer simulations of discrete-growth models [9] have so far given $\phi = 4$, while direct simulations of eq. (1) seem to give [5, 10] $\phi = 3$. If we assume that (9) remains valid for $\lambda(t) \sim t^{-\alpha}$, we find $W \sim t^{(4 - \alpha\phi)/12}$, if $\alpha < 1/\phi$. For $\phi = 4$ this agrees with our β in $d = 2$. In fact, we have checked the scaling form (9), with a time-dependent λ for *both* the GRSOS model *and* the KPZ equation, and indeed

obtained $\phi = 4$ for both models. This lends strong support to our direct determination of β (see fig. 2). To obtain this result for the KPZ equation, it is crucial to realize that the lattice constant of the mesh in which we solve the equation must be smaller than the length scale $v^3/\lambda^2 D$. For coarser discrete meshes, as used in the previous works [5,10], we observe a crossover to $\phi = 3$. Details of these results will be published elsewhere.

Experimentally, the phenomena described here can be probed with metastable systems such as a solid growing into a supercooled melt or into a supersaturated solution. The degree of metastability, which is proportional to λ , can be varied by, for example, progressively decreasing the degree of undercooling. Similarly, in a sputtered-growth experiment, our results describe a system as the sputtering rate is tuned to fall off algebraically in time. Thus we expect it to be experimentally feasible to probe the three properties we have discussed: the generalized relation of Galilean invariance, the possibility of a phase transition at physical dimensions, and the tunable roughening exponents.

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REFERENCES

- [1] KRUG J. and SPOHN H., in *Solids Far from Equilibrium: Growth, Morphology and Defects*, edited by C. GODRICHE (Cambridge University Press, New York, N.Y.) 1990.
- [2] KARDAR M., PARISI G. and ZHANG Y. C., *Phys. Rev. Lett.*, **56** (1986) 889.
- [3] FORSTER D., NELSON D. R. and STEPHEN M. J., *Phys. Rev. A*, **16** (1977) 732.
- [4] EDWARDS S. F. and WILKINSON D. R., *Proc. R. Soc. London, Ser. A*, **381** (1982) 17.
- [5] GROSSMANN B., GUO H. and GRANT M., *Phys. Rev. A*, **43** (1991) 1727.
- [6] HOHENBERG P. C. and HALPERIN B. I., *Rev. Mod. Phys.*, **49** (1977) 435.
- [7] LIPOWSKY R., in *Random Fluctuations and Pattern Growth: Experiments and Models*, edited by H. E. STANLEY and N. OSTROWSKY (Kluwer, Dordrecht, 1988).
- [8] KIM J. M. and KOSTERLITZ J. M., *Phys. Rev. Lett.*, **62** (1989) 2289; KIM J. M., KOSTERLITZ J. M. and ALA-NISSILA T., *J. Phys. A*, **24** (1991) 5569.
- [9] KRUG J., MEAKIN P. and HALPIN-HEALY T., *Phys. Rev. A*, **45** (1992) 638; AMAR J. G. and FAMILY F., *Phys. Rev. A*, **45** (1992) 5378.
- [10] FORREST B. M. and TORAL R., to be published in *J. Stat. Phys.* (1992).