Effect of long range interactions on the growth of compact clusters under deposition

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Abstract. In the paper the role of long range interactions on the growth of a volume conserving surface is studied using the Nonlocal Conserved Kardar-Parisi-Zhang (NCKPZ) equation. It is shown that previous theoretical predictions are inconsistent with an exact one-dimensional result. This serves as a motivation for construction of a Self-Consistent Expansion (SCE) that recovers the exact one-dimensional result, and gives the scaling exponents in higher dimensions as well. A possible application of this result to colloidal systems is discussed.

PACS. 05.70. Ln Nonequilibrium and irreversible thermodynamics – 02.50. -r Probability theory, stochastic processes, and statistics

Kinetic roughening of nonequilibrium interfaces is today a paradigmatic problem for theoretical tools in the field of nonequilibrium statistical physics and disordered systems, apart from being of practical importance. Therefore, it is not surprising that this phenomena has received much attention during the last two decades [1,2]. A common feature of many interfaces observed experimentally and in discrete growth models is that their roughening follows simple scaling laws [3]. The morphology and dynamics of a rough interface can be characterized by the surface width, W(L, t), that scales as

$$W\left(L,t\right) = \frac{1}{\sqrt{L}} \left\langle \sum_{\boldsymbol{r}} \left[h\left(\boldsymbol{r},t\right) - \bar{h}\left(t\right)\right]^{2} \right\rangle^{1/2} = L^{\alpha} f\left(\frac{t}{L^{z}}\right), \quad (1)$$

where $h(\mathbf{r}, t)$ describes the height of the interface above the point \mathbf{r} at time t and $\bar{h}(t)$ is the mean height of the interface at time t. α is the roughness exponent of the interface, z is the dynamic exponent that describes the scaling of the relaxation time with L — which is the size of the system. The brackets $\langle \cdots \rangle$ denotes noise averaging. The scaling function f(u) behaves like $f(u) \sim u^{1/z}$ for small u's (i.e. for $t \ll L^z$) and like a constant (i.e. $f(u) \sim \text{const}$) for large u's (i.e. for $t \gg L^z$). The scaling exponents α and z describe the asymptotic behavior of the growing interface in the hydrodynamic limit. Therefore, these quantities form the basic categories by which different models can be classified — namely universality classes.

The most prominent example for this scaling properties is, no doubt, the famous Kardar-Parisi-Zhang (KPZ) equation [2] that was formulated in order to describe a growing interface due to ballistic deposition. Actually, the KPZ equation is now considered to describe a very large class of different phenomena such as fluid flow in porous media, propagation of flame fronts, flux lines in superconductors not to mention deposition processes, bacterial growth and "DNA walk" [1]. In more technical terms, all these examples form the KPZ universality class.

The success of the KPZ equation in describing deposition phenomena motivated many researchers to develop a continuum growth model for volume conserving surfaces such as the technologically important molecular beam epitaxy (MBE) process [4–10]. The physical mechanism that distinguishes MBE from previously discussed growth processes is the surface diffusion of the deposited particles. It is well-known that in the temperature range of MBE growth, desorption of atoms and formation of overhangs and bulk defects is negligibly small. As a consequence the continuum model describing this processes must conserve the number of particles on the interface. The introduction of conservation laws into the growth equations forms new universality classes in surface phenomena, such as the one described by the Conserved Kardar-Parisi-Zhang (CKPZ) equation given by

$$\frac{\partial h\left(\boldsymbol{r},t\right)}{\partial t} = -K\nabla^{4}h\left(\boldsymbol{r},t\right) - \frac{\lambda}{2}\nabla^{2}\left(\nabla h\right)^{2} + \eta\left(\boldsymbol{r},t\right),\quad(2)$$

where K is a diffusion constant, λ is the coupling constant, and $\eta(\mathbf{r},t)$ is a conservative noise term (to be defined in Eq. (4) below). Still, the conserved KPZ model is not considered today as a realistic continuum model for MBE processes. The conserved KPZ model does not include the effects of step-edge barriers and the phenomenon of slope selection, that are experimentally well-known to dominate the MBE growth at long times. However, the MBE phenomenon will not be further discussed here for two reasons. First, as will be shown, already this simple model and its extensions elicit methodological difficulties

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that are explicitly addressed in this work. Second, there are other volume conserving growing surface whose properties can be described using this approach. One process of this kind is kinetic roughening in colloidal systems [14]. Yet, in that system one needs to account for the longrange hydrodynamic interactions that are known to exist [15,16] between the deposited particles and the surface below. Actually, even a simpler system of two colloidal particles diffusing in a fluid exhibit such long range interactions [17,18], with the difference that while for the two-body interaction explicit theoretical predictions can be made, for the interaction between a single particle and an interface below no prediction is available.

Interestingly, a model that captures both the longrange interactions and the mass conservations is known in the literature, namely the Nonlocal Conserved KPZ (NCKPZ) equation presented by [19,20]. The continuum equation for the height of the surface $h(\mathbf{r}, t)$ at a point \mathbf{r} and time t measured relative to its spatial average is

$$\frac{\partial h\left(\boldsymbol{r},t\right)}{\partial t} = -K\nabla^{4}h\left(\boldsymbol{r},t\right) - \frac{1}{2}\nabla^{2}\int d^{d}r'g\left(\boldsymbol{r}'\right) \\ \times \nabla h\left(\boldsymbol{r}+\boldsymbol{r}',t\right) \cdot \nabla h\left(\boldsymbol{r}-\boldsymbol{r}',t\right) + \eta\left(\boldsymbol{r},t\right), (3)$$

where K is a constant, the kernel $g(\mathbf{r})$ represents the long-range interactions with a short-range part $\lambda_0 \delta^d(\mathbf{r})$ and a long-range part $\sim \lambda_\rho r^{\rho-d}$, where d is the substrate dimension and ρ is an exponent characterizing the decay of the long range interaction. More precisely in Fourier space $\hat{g}(q) = \lambda_0 + \lambda_\rho q^{-\rho}$. Finally, $\eta(\mathbf{r}, t)$ is a conservative spatially correlated noise term that satisfies $\langle n(\mathbf{r}, t) \rangle = 0$

$$\langle \eta \left(\boldsymbol{r}, t \right) \eta \left(\boldsymbol{r}', t \right) \rangle \sim \nabla^2 \left| \boldsymbol{r} - \boldsymbol{r}' \right|^{2\sigma - d} \delta \left(t - t' \right),$$
 (4)

where σ is an exponent characterizing the decay of spatial correlations (in the case of conservative white noise , which is of great interest in physical systems, $\sigma = 0$).

In the next section I will present the current theoretical predictions for the scaling exponents of the NCKPZ model. It will become evident that all those results are inconsistent with an exact one-dimensional result obtained in the past. Then, in Section 3 the Self-Consistent Expansion (SCE) approach will be applied to this model. Interestingly, this method yields results that are consistent with the exact one-dimensional result. In addition, results for higher dimensions and for general ρ 's are presented. Eventually, a discussion and a summary are given in Section 4.

After suggesting the model, Jung et al. [19,20] investigated the NCKPZ problem using the Dynamic Renormalization Group (DRG) method [1], and derived a very rich and complex picture of phases — each one of them described by a different set of critical exponents. In this paper I will not go into the details of all those phases, but would like to focus on the strong coupling behavior of NCKPZ. In this regime the DRG analysis yields the power counting result

$$\alpha_{DRG} = \frac{2 - d - \rho + 2\sigma}{3} \qquad z_{DRG} = \frac{d + 10 - 2\rho - 2\sigma}{3},$$
(5)

where α is the roughness exponent and z is the dynamic exponent. This solution is said to valid for positive values of ρ and for $d < 2 + 2\rho + 2\sigma$.

Lately, another method, which proved useful in the context of local growth problems, was also applied to the NCKPZ problem. The method, is a Flory-type scaling approach that was originally proposed by Hentschel and Family [21] and generalized by Tang and Ma [22,23] to the nonlocal case. Interestingly, the authors claim that the DRG results presented in equation (5) above are just the weak-coupling exponents, while the strong-coupling dynamical exponents, obtained by using the scaling approach, are

$$\alpha_{SA} = \frac{2-\rho}{d+3-2\sigma} \qquad z_{SA} = 4-\rho - \frac{2-\rho}{d+3-2\sigma}.$$
 (6)

Surprisingly, both results do not agree with an exact result that is possible for some one-dimensional cases (namely for $\lambda_0 = 0$ and $\rho = 2 + 2\sigma$) of the NCKPZ problem. The exact result is obtained in reference [24] using the Fokker-Planck form associated with the Langevin form of NCKPZ equation given in equation (3) above. The exact values of the scaling exponents in one-dimension for $\lambda_0 = 0$ and $\rho = 2 + 2\sigma$ are

$$\alpha_{exact} = \frac{\rho - 1}{2} \qquad z_{exact} = \frac{9 - 3\rho}{2}.$$
 (7)

These exact results indicates that both methods (DRG and the scaling approach) are unreliable already in one dimension (not to mention higher dimensions). The inevitable conclusion is that in order to gain insight into the behavior of the NCKPZ model in any dimension, and for any value of ρ (the nonlocal parameter) and σ (the long-range noise parameter) it is necessary to employ a more reliable method.

The last point serves as another important motivation for this work. Namely, this model serves as a test case for the various theoretical tools that are available in this field. Thus, it might help to improve the current tools statistical physicists are using, so that more realistic and rich theories could be dealt with efficiently in the future.

In the following a method developed by Schwartz and Edwards [13,26,27] (also known as the Self-Consistent-Expansion (SCE) approach) is applied to the NCKPZ problem. This method has been previously applied successfully to the KPZ equation, and thus gained much credit by being able to give a sensible prediction for the KPZ critical exponents in the strong coupling phase, where, as is well-known, many Renormalization-Group (RG) approaches failed (as well as DRG of course) [28,29]. For the specific problem of NCKPZ, the SCE method indicates that two strong coupling solutions exist, one of which agrees with the power counting solution also obtained by DRG (presented in Eq. (5) above), while the other one recovers the exact one-dimensional result (and also extends it to higher dimensions). It turns out that each solution is valid for different values of the parameters σ , ρ and d (mutually excluding values).

The SCE method is based on going over from the Fourier transform of the equation in Langevin form, namely from

$$\frac{\partial h_q}{\partial t} = -Kq^4 h_q - \frac{\lambda_\rho q^{2-\rho}}{\sqrt{\Omega}} \sum_{\ell,m} \left(\boldsymbol{\ell} \cdot \boldsymbol{m}\right) \delta_{q,\ell+m} h_\ell h_m + \eta_q,\tag{8}$$

where η_q is the Fourier component of the noise that satisfies:

$$\langle \eta_q (t) \rangle = 0$$

$$\langle \eta_q (t) \eta_{q'} (t') \rangle = 2D_0 q^{2-2\sigma} \delta_{q+q'} \delta (t-t'), \qquad (9)$$

to a Fokker-Planck form, namely to

$$\frac{\partial P}{\partial t} + \sum_{q} \frac{\partial}{\partial h_{q}} \left[D_{0} q^{2-2\sigma} \frac{\partial}{\partial h_{-q}} + K q^{4} h_{q} + \frac{\lambda_{\rho} q^{2-\rho}}{\sqrt{\Omega}} \sum_{\ell,m} \left(\boldsymbol{\ell} \cdot \boldsymbol{m}\right) \delta_{q,\ell+m} h_{\ell} h_{m} \right] P = 0, (10)$$

(where $P(\{h_q\}, t)$ is the probability of having the configuration $\{h_q\}$ at a specific time t). and constructing a self-consistent expansion of the distribution of the height field (namely an expansion for $P(\{h_q\}, t)$).

The expansion is formulated in terms of ϕ_q and ω_q , where ϕ_q is the two-point function in momentum space, defined by $\phi_q = \langle h_q h_{-q} \rangle_S$, (the subscript *S* denotes steady state averaging), and ω_q is the characteristic frequency associated with h_q . It is expected that for small enough *q*'s, ϕ_q and ω_q obey the power laws in *q*

$$\phi_q = Aq^{-\Gamma}$$
 and $\omega_q = Bq^z$, (11)

where z is just the dynamic exponent, and the exponent Γ is related to the roughness exponent α by

$$\alpha = \frac{\Gamma - d}{2}.\tag{12}$$

The SCE method produces, to second order in this expansion, two nonlinear coupled integral equations in ϕ_q and ω_q , that can be solved exactly in the asymptotic small q limit to yield the required scaling exponents governing the steady state behavior and the time evolution.

the steady state behavior and the time evolution. By defining $M_{q\ell m} \equiv \frac{\lambda_p}{\sqrt{\Omega}} q^{2-\rho} (\boldsymbol{\ell} \cdot \boldsymbol{m}) \, \delta_{q,\ell+m}, K_q = Kq^4$ and $D_q = D_0 q^{2-2\sigma}$ it can be seen that the NCKPZ equation is of the general form discussed in references [26,27], where the SCE method is derived. Thus, the two coupled non-linear integral equations can be obtained easily, and read

$$D_{q} - K_{q}\phi_{q} + 2\sum_{\ell,m} \frac{M_{q\ell m}M_{q\ell m}\phi_{\ell}\phi_{m}}{\omega_{q} + \omega_{\ell} + \omega_{m}} - 2\sum_{\ell,m} \frac{M_{q\ell m}M_{\ell m q}\phi_{m}\phi_{q}}{\omega_{q} + \omega_{\ell} + \omega_{m}} - 2\sum_{\ell,m} \frac{M_{q\ell m}M_{m\ell q}\phi_{\ell}\phi_{q}}{\omega_{q} + \omega_{\ell} + \omega_{m}} = 0$$
(13)

and

$$K_q - \omega_q - 2\sum_{\ell,m} M_{q\ell m} \frac{M_{\ell m q} \phi_m + M_{m\ell q} \phi_\ell}{\omega_\ell + \omega_m} = 0, \quad (14)$$

where in deriving the last equation the Herring consistency equation [30] is used. In fact Herring's definition of ω_q is one of many possibilities, each leading to a different consistency equation. But it can be shown, as previously done in reference [27], that this does not affect the exponents (universality).

A full solution of equations (13) and (14) in the limit of small q's (i.e. large scales) yields a very rich family of solutions that will not interest us here, but may be the subject of a future detailed paper. Instead, I focus on the strong coupling solutions obtained by SCE. As mentioned above, two strong coupling solutions are obtained by SCE (they are strong coupling solutions in the sense that they do not exist when the coupling λ is set to zero), each of them describes the critical exponents for a different set of the parameters ρ , σ and d.

The first solution is obtained just by power counting, and reads $z = (d+10-2\rho-2\sigma)/3$ and $\Gamma = (d+4-2\rho+4\sigma)/3$. This solution is valid for $\rho-1 < \sigma$, $d < 6-2\sigma + \rho$, $d < 2+2\sigma+2\rho$ and $d < 8+2\sigma-4\rho$. Therefore, by direct inspection, this solution is not valid for the one-dimensional case with $\rho = 2 + 2\sigma$, and hence does not and is not expected to recover the exact Gaussian result obtained in reference [24]. It should also be mentioned that when translated to frequently used notation (using Eq. (12)) it can be seen that this solution is actually the same as the one obtained by the DRG analysis as in equation (5). In that sense the SCE approach agrees with the DRG analysis in that this solution is a strong-coupling one, and not a weak-coupling solution as claimed in references [22,23].

The second solution is a non power counting solution. It turns out that for d = 1 equation (13) is exactly solvable, and yields $\Gamma = \rho$. This result is actually the exact Gaussian solution of the one-dimensional case, mentioned above. In addition, from equation (14), the dynamical exponent can be extracted for a certain range of the parameters, namely for $1 < \rho < 3$ and $\rho > (4\sigma + 5)$, where it becomes $z = (9 - 3\rho)/2$.

For dimensionality higher than one (i.e. $d \ge 2$) such an exact solution in closed form cannot be found, and the second strong coupling solution is determined from the combination of the scaling relation $z = (d + 8 - \Gamma - 2\rho)/2$, and the transcendental equation $F(\Gamma, z, \rho) = 0$, where F is given by

$$F(\Gamma, z, \rho) = -\int d^{d}t \frac{\boldsymbol{t} \cdot (\hat{\boldsymbol{e}} - \boldsymbol{t})}{t^{z} + |\hat{\boldsymbol{e}} - \boldsymbol{t}|^{z} + 1} \left[(\hat{\boldsymbol{e}} \cdot \boldsymbol{t}) |\hat{\boldsymbol{e}} - \boldsymbol{t}|^{2 - \rho} t^{-\Gamma} \right]$$
$$+ \hat{\boldsymbol{e}} \cdot (\hat{\boldsymbol{e}} - \boldsymbol{t}) t^{2 - \rho} |\hat{\boldsymbol{e}} - \boldsymbol{t}|^{-\Gamma} \right] + \int d^{d}t \frac{\left[\boldsymbol{t} \cdot (\hat{\boldsymbol{e}} - \boldsymbol{t}) \right]^{2}}{t^{z} + |\hat{\boldsymbol{e}} - \boldsymbol{t}|^{z} + 1} t^{-\Gamma} |\hat{\boldsymbol{e}} - \boldsymbol{t}|^{-\Gamma},$$
(15)

and \hat{e} is a unit vector in an arbitrary direction. The last equation has to be solved numerically of course. In addition, this solution is valid as long as the solutions of the last equations satisfy the following four conditions: $d + 4 - 3\Gamma - 2\rho + 4\sigma < 0, \ d - 3\Gamma + 2\rho < 0, \ d < \Gamma$ and $d - 4 - \Gamma + 2\rho < 0$.

It should be mentioned that SCE yields many other "weak coupling" solutions to NCKPZ. They are called "weak coupling" in the sense that they can be obtained from a linear conserved growth problem with correlated noise. These solutions are also not obtained by DRG nor by the Scaling Approach.

In summary, in this paper two theoretical predictions for the strong coupling phase of the NCKPZ model using DRG and a scaling approach were presented. It was shown that these results are inconsistent with a recent exact one-dimensional result obtained in reference [24]. This discrepancy served as a motivation for the construction of a Self Consistent Expansion (SCE) that made possible the calculation of the critical exponents for the most general case (i.e. any ρ , σ and d). I obtained two possible strong coupling solutions. The first strong coupling solution is a power counting one, and is identical to the previous result of the DRG analysis (actually it was also obtained by the Scaling Approach — however, it was mistakenly identified as a weak coupling one). The second strong coupling solution is a non power counting solution, and is not obtained by DRG nor by the Scaling Approach. For d = 1, this solution turns out to be the exact Gaussian solution obtained in reference [24].

In the context of roughening of colloidal systems, as the interactions between a falling colloidal particle and an interface is not known analytically, the results presented here can useful in their systematic study. To begin with, one can easily determine the range of the interactions by measuring the roughness. One can also study the influence of the geometry of the sample, the dimensionality, and the possible electrostatic interaction between the colloidal particles. Taking another direction, as equation (3) reproduces some of the measured features in a solid manner, it could serve as an inspiration for a more rigorous evaluation of the hydrodynamical interactions. It is more than likely that the real interaction is much more complicated than equation (3), but it can be the case that it captures some leading order behavior such that subleading terms are irrelevant in the RG sense [1]. An interesting option is that once one determines the range of the interactions, namely the parameter ρ in equation (3), from either experimental, numerical or analytical results, more detailed properties could be obtained directly from the equation.

Coming back to the Dynamic Renormalization Group approach, the reasons for its failure in recovering the exact one-dimensional results for the nonlocal models are not fully clear. It seems that DRG is not able to calculate the strong-coupling solution because the propagator of the linear theory $G_0(q, \omega)$ that is used in DRG in the perturbative expansion is not a good candidate. If instead an expansion is made around a different free model better results can be achieved. It is suspected that the nonlinear term generates a fractional relaxation term (namely a fractional biharmonic operator) under renormalization. Thus, the propagator should be modified. Actually, this idea is implemented in the Self-Consistent Expansion. Therefore, an application of that method to the exactly solvable model described above, indeed reproduces the exact results. This suggests that there is an advantage in using the SCE to deal with such nonlinear Langevin equations.

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