Pyramid Growth Without Deposition Noise

D. C. Vernon, M. Siegert, and M. Plischke
Department of Physics, Simon Fraser University, Burnaby, B.C. Canada V5A 1S6
(April 16, 1999)

Several models of molecular beam epitaxy, both atomistic and ones based on Langevin equations, have as one of their generic growth scenarios the formation of three-dimensional structures such as mounds or pyramids. The characteristic size R of these structures increases as a function of deposition time with a power law $R \sim t^n$. In order to investigate the dependence of the growth exponent n on the characteristics of the fluctuations of the deposition flux we compare results of Monte-Carlo simulations for random deposition and for deposition on an artificially constructed deterministic sequence of sites. Although the latter algorithm leads to much smaller height fluctuations on each site, the growth exponent in both cases is found to be close to 0.25.

PACS number(s): 05.70.Ln, 68.55.-a, 81.10.Aj

I. INTRODUCTION

Experimental work in molecular beam epitaxy has shown a variety of different growth modes, including the formation of three-dimensional structures on initially flat surfaces. This paper deals with one of these modes: the formation of pyramids in homoepitaxial growth. Experiments on different systems have shown that three-dimensional structures occur only for some material parameters and surface orientations, and can produce mounds or pyramids[1–7]. The systems which do show three-dimensional growth have as a common feature a characteristic size of the structures R, which grows as $R \sim t^n$, with n between[3–7] 0.16 and 0.33.

The microscopic mechanism that gives rise to this three-dimensional growth mode is generally believed to be a potential energy barrier at step edges[8,9] that particles diffusing on a terrace must cross in order to reach a lower terrace. Such a diffusion bias[10] results in a positive or uphill contribution to the adatom diffusion current that causes the initially flat high-symmetry surface orientation to become unstable[10,11].

Other effects, such as the transient mobility[12,13] of newly deposited atoms and effects due to crystalline symmetries of the growing film[14], give rise to a negative contribution to the surface current. At sufficiently large slopes these contributions can perfectly balance the current due to the step-edge barriers[15]. At these characteristic slopes the net surface current is zero and the corresponding surface orientations are stable[14–16]. This phenomenon is commonly called slope selection.

As soon as pyramids or mounds with side planes corresponding to the characteristic slopes have developed the average magnitude of the slope remains approximately constant. However, the pyramids and mound formations coarsen as more and more material is deposited: smaller pyramids disappear and larger pyramids grow. This coarsening phenomenon has been the subject of several recent publications[13–15,17–24] and is also the topic of this article.

II. LANGEVIN DESCRIPTION

Within a continuum theory the evolution of surface morphology can be described by a Langevin equation of the form

$$\frac{\partial h}{\partial t} = -D\Delta\Delta h - \nabla \cdot \mathbf{j}(\mathbf{m}) + \eta \tag{1}$$

where $\eta(\mathbf{r}, t)$ is random noise and $\mathbf{j}$ is the nonequilibrium surface current, which is a function of the local slope $\mathbf{m}(\mathbf{r}, t) = \nabla h(\mathbf{r}, t)$. Terms that break the $h \to -h$ symmetry[10,25] may be added to the right-hand side of Eq. (1) as well. However, results obtained by Politi[23] show that such terms, at least in one dimension, do not change the growth law; we therefore omit such contributions to the surface current for the sake of simplicity.

The form of the nonequilibrium surface current results from a combination of different effects. Diffusion bias leads[10,11,14] for small slopes m to a current that is proportional to the slope, $\mathbf{j}_b = c\mathbf{m}$ with $c > 0$. This causes the high-symmetry orientation $\mathbf{m} = 0$ to become unstable. For larger slopes m in the step-flow regime this current $j_b \simeq F/m$. Both regimes can be described (at least in one dimension) within the Burton-Cabrera-Frank theory[26]. Transient mobility effects give rise to a contribution[15] $j_{\rm tm} = -p^2 F m$. The coefficient p is usually of order one leading to a zero in the net current $\mathbf{j} = \mathbf{j}_b + \mathbf{j}_{\rm tm}$ in the step-flow regime at $m_0 \simeq 1/p$. Crystalline anisotropies of the growing film ensure that there is only a finite number of such stable orientations[14,24]. In the framework of the Langevin description all this is taken into account by choosing an appropriate interpolation formula[6,14,15,18,21,24] for the surface current $\mathbf{j}(\mathbf{m})$.

It is important to note, however, that although the relative size of these different mechanisms determines the *direction* of the slope-dependent current, the very existence of such a current is due to deposition. In the absence of deposition, detailed balance guarantees that there is no slope-dependent current. Therefore, in experiments the evolution of the surface morphology is necessarily influenced by fluctuations of the deposition flux corresponding to a nonzero noise strength η. In principle, thermal noise $\nabla \cdot \boldsymbol{\eta}_{\mathrm{th}}$ due to the stochastic nature of surface diffusion should also be added to the right-hand side of Eq. (1). This is commonly omitted because the long-wavelength modes are believed to dominate the evolution of surface morphology for sufficiently late times. Since the spectrum of $\nabla \cdot \boldsymbol{\eta}_{\mathrm{th}}$ is smaller by a power of $\mathbf{k}$ than that of η, thermal fluctuations are usually neglected in the Langevin approach.

III. EFFECTS OF THE NOISE

Numerical integrations of the Langevin equation (1) have generally found that the characteristic size R of pyramids or mounds increases like a power law[14,18,27,24], $R \sim t^n$ with typically $n \lesssim 0.25$. Most of these integrations were done for zero noise $\eta = 0$ following the common belief that noise is irrelevant for such growth processes[15]. This is confirmed in numerical integrations of Langevin equations with nonzero noise, where within the numerical uncertainties no difference in the growth exponent could be found[18].

Recently Tang *et al.* (TSV)[22] have proposed that the deposition noise η can play a dominant role in the coarsening dynamics of pyramids. For uncorrelated noise the number of particles falling into any given region of the surface is a random walk, independent of any other region. This causes the mass fluctuations to grow with the square root of the number of particles deposited. From this TSV deduce the power law $R \sim t^{1/4}$, i.e., $n = 1/4$. In their picture, the current $\mathbf{j}(\mathbf{m})$ determines the slope of the pyramids and also plays a role in the initial development of three-dimensional structure, but does not contribute to the growth of these structures once they reach their steady-state shape.

The above argument relies on the assumptions that there is single length scale that dominates the coarsening problem, and that the surface performs a random walk due to the fluctuations of the flux. Both assumptions are highly questionable[24]: The dynamics are governed by at least two different length scales with different time dependences, and the coarsening is hampered by metastable states that tend to trap the evolution, if several adjacent pyramids are of roughly the same size. However, the question of whether or not stochastic fluctuations play an important role for the coarsening dynamics remains relevant, if rephrased in the following way: Is it possible that such fluctuations provide a mechanism to overcome the barriers in such metastable states? In a Langevin description this question can be answered by varying the strength of the noise η. In Fig. 1 we show the results of numerical integrations of Eq. (1) with a surface current[14,15,24] with components $j_{x,y} = m_{x,y}(1-m_{x,y}^2)$ for different noise strengths ϵ defined via the noise correlation $\langle \eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle = 2\epsilon\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$. There are two noticeable effects of the noise that both lead to a larger

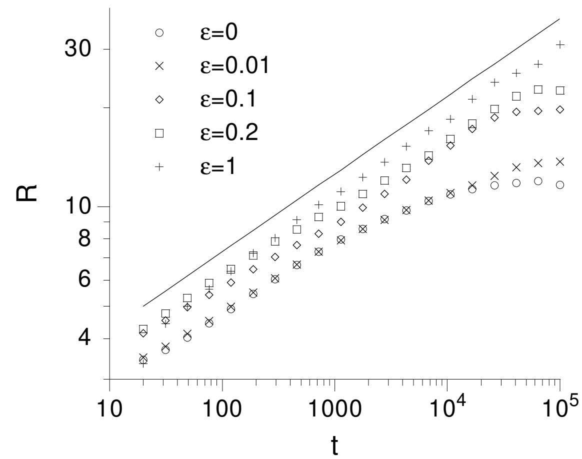

FIG. 1. Numerical integration of the Langevin Eq. (1) for different noise strengths ϵ. In all cases the system size $L = 192$ is much larger than the pyramid size R at the latest time so that finite size effects due to the periodic boundary conditions could not be observed. The solid line indicates a power law $R(t) \sim t^n$ with $n = 1/4$.

pyramid size for increasing noise strengths after a fixed deposition time. Firstly, the initial instability develops faster with stronger fluctuations. This leads to a parallel shift of the data to larger pyramid sizes for larger ϵ that is easily visible in Fig. 1. Secondly, for all noise strengths ϵ that are shown in Fig. 1 except for $\epsilon = 1$ the pyramid size $R(t)$ saturates. This asymptotic value $R_\infty = R(t \to \infty)$ also increases with ϵ. As explained in Ref. 24 the growth of pyramids is hampered by metastable states. When the system becomes trapped in such a state, the coarsening stops until fluctuations drive the system over the barriers of that metastable state. Since these barriers increase linearly with the pyramid size,[24] a larger R_∞ requires a larger noise strength ϵ. Conversely, for any fixed noise strength the pyramid size $R(t)$ will become so large that the noise fluctuations are insufficient to drive the system out of such metastable states, i.e., even for the noise strength $\epsilon = 1$ it is expected that coarsening will cease at some later time. For times t such that $R(t) \ll R_\infty$ the coarsening is well described by a power law $R(t) \sim t^{1/4}$ as indicated by the solid line[24]. All these results lead to the conclusion that in a continuum description noise is indeed irrelevant for the coarsening dynamics. It only affects the initial pyramid size and the time to reach the asymptotic size R_∞.

IV. MONTE-CARLO SIMULATIONS WITH DETERMINISTIC DEPOSITION

In the remainder of this article we show how this problem can be investigated by means of Monte-Carlo simulations. We used the same dynamics as in previous work[18] to describe the deposition and diffusion of particles on a growing surface. The surface is described by a column of particles of height $h(x,y,t)$ at each lattice site on a square lattice, with additional particles dropped from above. This solid-on-solid model is intended to describe growth without voids or overhangs, which are usually not observed in molecular beam epitaxy.

To simulate the effect of transient mobility and to produce a downward component of the nonequilibrium diffusion current, we allow each newly deposited particle to immediately attempt one further move. If one or more of the nearest neighbour columns of the deposition site are at *lower* height, the deposited particle hops to one of them with a probability p=0.1[18].

After incorporation, all particles that are not fully buried are allowed to diffuse by hopping to the top of one of the nearest neighbour columns. This diffusion process is controlled by a local energy function and obeys detailed balance. Thus, in the absence of deposition, the surface relaxes to its equilibrium state (faceted or rough). The energy of a particle at the top of the column at $\mathbf{r}$ is composed of two terms

$$E(\mathbf{r}) = -E_1 n_1(\mathbf{r}) - E_b n_2(\mathbf{r}) \,, \tag{2}$$

where $n_1(\mathbf{r})$ is the number of lateral nearest neighbours of the particle at $\mathbf{r}$ and $n_2(\mathbf{r})$ is the number of particles in the neighbouring columns at heights $h(\mathbf{r}) \pm 1$. The hopping rates are chosen so as to model the activated nature of surface diffusion and to produce diffusion bias. We take

$$W\left[h(\mathbf{r}) \to h(\mathbf{r}) - 1, h(\mathbf{r}') \to h(\mathbf{r}') + 1\right] = W_1 W_b \tag{3}$$

where

$$W_1\left[n(\mathbf{r})\right] = \frac{1}{\tau_{dif}} \exp\left[-\frac{E_1}{k_b T} n_1(\mathbf{r})\right] \tag{4}$$

and

$$W_b[n_2(\mathbf{r}') - n_2(\mathbf{r})] = \exp\left[\frac{E_b}{k_B T}\left(n_2(\mathbf{r}') - n_2(\mathbf{r})\right)\right] \tag{5}$$

for $n_2(\mathbf{r}') < n_2(\mathbf{r})$, and 1 otherwise. The first part W_1 depends only on the configuration of the particle at site $\mathbf{r}$ and is an Arrhenius like activated process. The second piece, W_b, produces diffusion bias: When a particle attempts to hop to the edge of a terrace from the upward side, it is required to cross a barrier of energy E_b and this process is therefore suppressed. The parameters in the above rates were chosen to put the zero in the current at a reasonable slope and to minimize cross-over effects. See Ref. 18 for a discussion of how the appropriate parameters were determined.

In order to investigate the effects of fluctuations particles are no longer deposited randomly. Instead, they are deposited in a pattern with much smaller fluctuations in the number of particles in a given area. In fact, the size of the fluctuations is bounded, and can reach zero for some times. The patterns chosen are based on Sobol' sequences[28], which are used for numerical integration. These sequences are chosen to uniformly cover an area, without the fluctuations in number that are associated with a random set of points. The sequences also cover the area in a "well spread out" fashion: later points fill in the areas between earlier ones. The points are not placed on a fixed grid, so there is no built-in length scale, and they are distributed evenly over the surface, so there is no imposed current. There are many different possible Sobol' sequences, each defined by a set of binary fractions called *direction numbers*, and by a few initializing points. Each number in a Sobol' sequence is generated from the previous one by a bitwise exclusive or with one of these direction numbers, chosen to produce binary fractions with slowly increasing denominators. See Press *et al.*[28] for a description of the algorithm used to generate these sequences, and the version of the Sobol' sequence used, the Sobol'-Antonov-Saleev sequence. If two of these sequences are used to choose the lattice sites for deposition, the fluctuation in the height at any site is small and bounded (see Fig. 2).

For deposition in a Sobol' sequence, the structures that result are similar to those for random deposition. Since the deposition is much more uniform, the Sobol' case produces more regular structures, as the random case does for smaller deposition rates. See Fig. 3 for an example of the surface morphology that does appear. The fluctuations that obscure pyramid formation for the same deposition rate in the random case, and in experiments at high deposition rates and/or low temperatures, do not

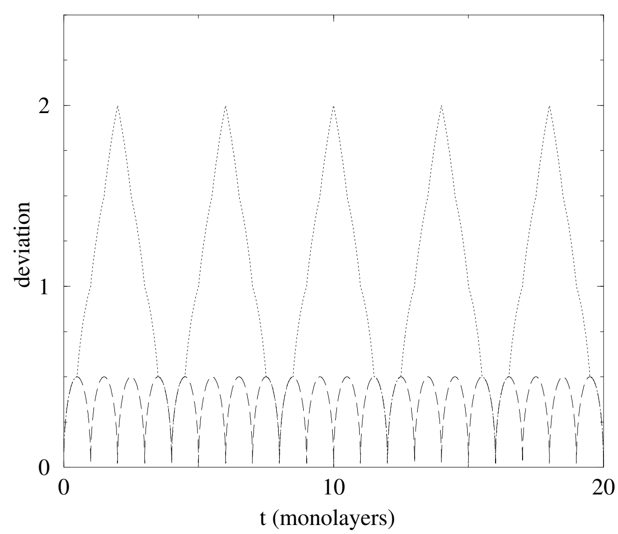

FIG. 2. Root mean square deviation of the height at each lattice site of a $L^2 = 64^2$ lattice, for deposition only, in two different Sobol' sequences (dashed and dotted lines, respectively). The two sequences are defined by different sets of direction numbers. The sequence corresponding to the dashed line is constructed such that all lattice sites are visited before any site in the next layer is filled, whereas for the other sequence it takes four layers until a perfectly flat surface is reestablished.

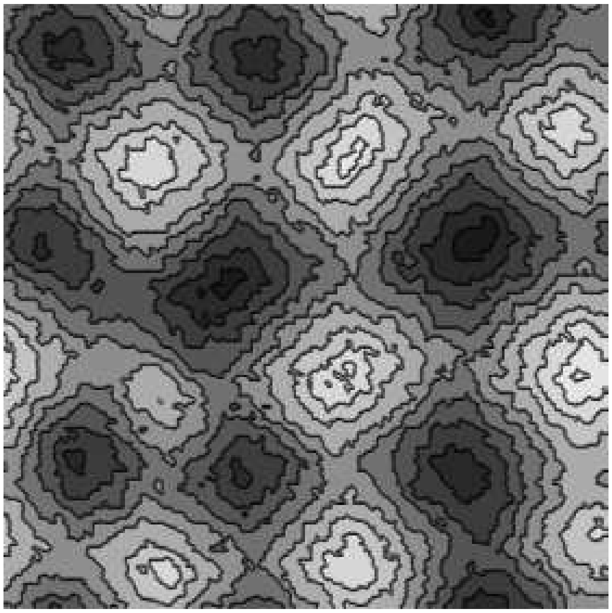

FIG. 3. Typical structure for a 128^2 lattice after 10^4 monolayers deposited, for Sobol' deposition. The height differences range from -10 (black) to $+10$ (white) monolayers.

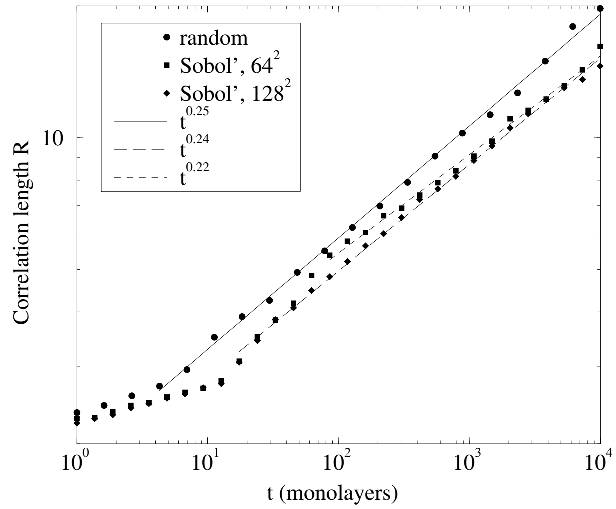

FIG. 4. Time dependence of the first zero of the height-height correlation function, for both random deposition and deposition in a Sobol' sequence, for 64^2 and 128^2 surfaces, and a deposition rate of 10^{-2} depositions/diffusion moves.

occur. This makes it clear that deposition drives an uphill current, which causes three-dimensional structures to appear. Random fluctuations are not necessary to trigger the instability.

To determine the growth law for the size of the structure on the surface, the slope-slope correlation function $C(\mathbf{r},t) = \langle \mathbf{m}(\mathbf{0},t)\mathbf{m}(\mathbf{r},t)\rangle$ was calculated, and its first zero used as a measure of the size of domains. For both random deposition and deposition in a Sobol' sequence, the growth obeyed a power law for large times, with exponent $n \approx 0.25$ for the random case and $n \approx 0.22$ for Sobol' deposition on a 64^2 lattice and $n \approx 0.24$ for a 128^2 lattice, as shown in Fig. 4. The difference does not seem to be significant and is certainly less than any reasonable estimate of the uncertainty in either n. The behaviour at short times is clearly different in the case of Sobol' deposition, as might be expected. It takes longer for the more uniform Sobol' sequence to deposit enough particles in one place to destabilize the initial high-symmetry orientation of the surface, and to enter the three-dimensional growth regime. This difference in the initial behaviour is the only significant difference that allows us to differentiate between random and deterministic deposition in our simulations. The deposition rate used in these simulations is relatively large (in fact, if compared to experiments, unrealistically large). However, in the case of Sobol' deposition the integral over time of the fluctuations in the deposition rate is bounded. Therefore, any noise-driven coarsening must stop when the characteristic length scale becomes large enough. If there were any contribution to the coarsening from such a mechanism one would expect to see a cross over at this characteristic length scale. This was not observed in our simulations.

In all our Monte-Carlo simulations, with random or deterministic deposition, we were not able to observe a slowing of the coarsening rate due to metastable states. This is almost certainly due to the high deposition rates used in our simulations. Even for the deterministic deposition using Sobol' sequences the evolving morphologies as shown in Fig. 3 are far more random and rougher than the morphologies obtained in numerical integrations of Langevin equations corresponding to the late times of Fig. 1. Currently we are unable to simulate substantially smaller deposition rates because of the enormous amount of necessary computer time. However, using different techniques[29] it should be possible to detect these effects of metastable states at small deposition rates also in a Monte-Carlo simulation. It should also be noted that thermal noise $\boldsymbol{\eta}_{\mathrm{th}}$ is inevitably present in Monte-Carlo simulations, and this may also help to overcome barriers between states.

V. CONCLUSION

The coarsening behaviour of three-dimensional growth in molecular beam homoepitaxy was studied by numerical integrations of Langevin equations with and without noise and by Monte-Carlo simulations of a microscopic model with random or deterministic deposition. In the case of the Langevin equation, the breaking of detailed balance that is necessary for the slope-dependent surface currents to occur is put in by hand ($j(\mathbf{m})$ in (1)). The presented Monte-Carlo simulations show that, as expected, even quite regular irreversible deposition is sufficient to break detailed balance and to set in motion the deterministic part of the coarsening process.

Our results obtained from numerical integrations of

Langevin equations as well as from Monte-Carlo simulations show that white noise in the deposition process leads to coarsening behaviour that is indistinguishable from the behaviour found for deterministic deposition. Only the initial regime that describes the onset of the instability is different. In the case of Langevin dynamics it is found that coarsening slows down and stops at late times even for nonzero noise strength. All these results are in agreement with a recent theory[24] which explains that the coarsening exponent $n \approx 1/4$ is due to the interactions between the topological defects in the system.

This research was supported by the NSERC of Canada.

---

[1] G. W. Smith, A. J. Pidduck, C. R. Whitehouse, J. L. Glasper, and J. Spoward, J. Cryst. Growth **127**, 966 (1993).

[2] C. Orme, M. D. Johnson, K. T. Leung and B. G. Orr, in *Compound Semiconductor Epitaxy*, edited by C. W. Tu, L. A. Kolodziejski and V. R. McCrary, MRS Symposia No. 340 (Materials Research Society, Pittsburgh, 1994), p. 233.

[3] H.-J. Ernst, F. Fabre, R. Folkerts, and J. Lapujoulade, Phys. Rev. Lett. **72**, 112 (1994).

[4] K. Thürmer, R. Koch, M. Weber, and K. H. Rieder, Phys. Rev. Lett. **75**, 1767 (1995).

[5] J.-K. Zuo and J. F. Wendelken, Phys. Rev. Lett. **78**, 2791, (1997).

[6] J. A. Stroscio, D. T. Pierce, M. D. Stiles, and A. Zangwill, Phys. Rev. Lett. **75**, 4246 (1995).

[7] F. Tsui, J. Wellmann, C. Uher, and R. Clarke, Phys. Rev. Lett. **76**, 3164 (1996).

[8] G. Ehrlich and F. G. Hudda, J. Chem. Phys. **44**, 1039 (1966); S. C. Wang and G. Ehrlich, Phys. Rev. Lett. **70**, 41 (1993).

[9] R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. **37**, 3682 (1966); R. L. Schwoebel, J. Appl. Phys. **40**, 614 (1969).

[10] J. Villain, J. Phys I (France) I **1**, 19 (1991).

[11] M. D. Johnson, C. Orme, A. W. Hunt, D. Graff, J. Sudijono, L. M. Sander, and B. G. Orr, Phys. Rev. Lett. **72**, 116 (1994).

[12] M. C. Bartelt and J. Evans, Surf. Sci. **298**, 421 (1993); J. W. Evans, Phys. Rev. B **43**, 3897 (1991); H. C. Kang and J. W. Evans, Surf. Sci. **269-270**, 784 (1992).

[13] M. C. Bartelt and J. Evans, Phys. Rev. Lett. **75**, 4250 (1995).

[14] M. Siegert and M. Plischke, Phys. Rev. Lett. **73**, 1517 (1994).

[15] M. Siegert in *Scale Invariance, Interfaces, and Non-Equilibrium Dynamics*, edited by A. J. McKane, M. Droz, J. Vannimenus, and D. Wolf, NATO ASI Series B, Vol. 344 (Plenum, New York, 1995), pp. 165-202.

[16] J. Krug, M. Plischke, and M. Siegert, Phys. Rev. Lett. **70**, 3271 (1993).

[17] P. Šmilauer and D. D. Vvedenski, Phys. Rev. B **52**, 14263 (1995).

[18] M. Siegert and M. Plischke, Phys. Rev. E **53**, 307 (1996).

[19] P. Politi and J. Villain, Phys. Rev. B **54**, 5114 (1996).

[20] J. Amar and F. Family, Phys. Rev. B **54**, 14742 (1996).

[21] M. Rost and J. Krug, Phys. Rev. E **55**, 3952 (1997).

[22] L.-H. Tang, P. Šmilauer, and D. D. Vvedensky, Eur. Phys. J. B **2**, 409 (1998); L.-H. Tang, Physica A **254**, 135 (1998).

[23] P. Politi, Phys. Rev. E **58**, 281 (1998).

[24] M. Siegert, Phys. Rev. Lett. **81**, 5481 (1998).

[25] T. Sun, H. Guo, and M. Grant, Phys. Rev. A **40**, 6763 (1989).

[26] J. Krug and M. Schimschak, J. Phys. I (France) **5**, 1065 (1995).

[27] M. Siegert, Physica A **239**, 420 (1997).

[28] W. H. Press, S. A. Teukolsky, W. T. Vetterling, and B. R. Flannery, *Numerical Recipies in C*, 2nd ed., (Cambridge University Press, Cambridge, 1992).

[29] A. B. Bortz, M. H. Kalos, J. L. Lebowitz, J. Comp. Phys. **17**, 10 (1975).