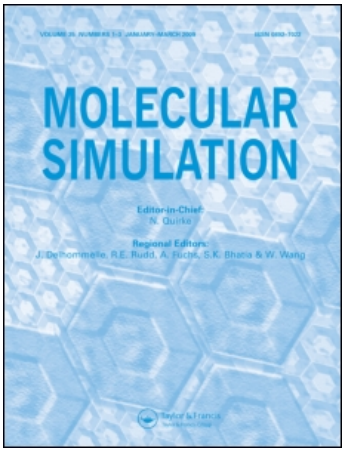

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The Universality Concept as a Tool to Simplify Numerical Simulations

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A number of physical systems exhibit naturally self-similar features, characterized by power-law correlations over some spatial and/or temporal domain. Critical or self-organized critical systems are general examples, and they appear in all fields of physics dealing with *N* interacting constituents. The powerful renormalization-group theory is able to answer many precise questions concerning these systems. But by for the most important consequence of the theory is the concept of the universality class. This fundamental concept can be used to simplify dramatically the numerical simulations of such systems, leading to the exact values of the relevant physical quantities—those who are related to the long-range correlations. In this paper, we develop the general method with the practical steps to apply it to concrete physical situations. We then focus on the particular case of the aggregates of colloidal particles.

Keywords: Self-similarity; Colloidal aggregates; Fractals; Critical systems

SELF-SIMILAR SYSTEMS

Among the physical systems of a large number, say N, of constituents in interaction, one can distinguish the very particular ensemble of the self-similar systems [1]. We shall discuss in this paper only spatial self-similarity, but temporal self-similarity exists too-for example the dynamics of sand-pile avalanches [2], and can be treated in the similar way. Self-similarity means that correlations follow power-laws, in contrast with the usual exponential decrease for the liquid phase, or the constant correlations for the crystal phase. The proper sense of the word "self-similarity" refers to the invariance of the system with respect to change of scale. In short, any piece of the system, after magnification, looks similar to the whole. In the context of geometry, they are called fractals [3]. But in the context of physics, it is more suitable to use the term 'self-similar', or scale-invariant, in order to enhance the physical feature that, when analyzed with probes of various length scales, the system show similar results. Nevertheless, in reality two natural cut-offs for self-similarity have to be considered: one certainly cannot consider pieces smaller than the typical size of the individual constituents, nor larger than the overall size of the systems.

The critical systems are the most famous examples. The Ising spin system at the critical temperature is the basic model of this kind: the spatial distribution of the spins "up" (respectively "down") is self-similar [4]. Note that for these critical systems, one has to tune very precisely the control parameter (e.g. the temperature, for the Ising spin magnetic system) in order to get the system exactly at the critical point. In contrast, there is a vast class of system which are at the critical state without fine-tuning any parameter. These are the self-organized critical systems [5]. Some are very simple: for example, the track of a Brownian particle in the 3-dimensional space is fractal, with fractal dimension 2. This means for example that the 2-point correlation function (corresponding to the probability that the diffusing particle passes through two locations r apart) decreases with distance r, as $1/r$. Other self-organized systems are less simple, as the aggregates of colloidal particles, according to the physico-chemical conditions, nanometric particles in a fluid can aggregate sequentially. They then form fractal aggregates with few very particular values of the fractal dimension [6]. We discuss here one way to model such a system

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and how to find the exact values of the fractal dimension from efficient numerical simulations.

FUNDAMENTAL CONSEQUENCES OF THE RENORMALIZATION-GROUP THEORY

One remarkable tool to study theoretically the self-similar systems is the renormalization-group theory [7]. To give a brief description, it is based on the idea of self-invariance one redefines locally the constituents of the system in larger units, by grouping them together (i.e. modifying the length scale), then redefining the interactions between the new units in order to get a similar system at another length scale. Reproducing indefinitely this process, one obtains a self-similar system where only the parameters linked to the self-invariant morphology are kept. One crucial consequence is the splitting of the set of all the physical parameters of the system into two ensembles: the list of the parameters which go to a finite non-trivial value after the renormalization scheme, and the list of the parameters which vanish. The former ensemble is the set of the relevant parameters, and the latter is the set of the irrelevant ones.

The interpretation of this result is that the proper value of any irrelevant parameter does not even remotely influence the self-similar structure of the system as a whole, provided the system is infinite. They can be set from the beginning to any trivial value (usually 0), and the resulting system, at the end, will be exactly the same. This is the idea which will be developed now in the context of the numerical simulations.

SIMPLIFICATION OF NUMERICAL SIMULATIONS OF SELF-SIMILAR SYSTEMS

The general simplification scheme is as follows. Suppose you have to model a system of N constituents, interacting through some complicated interactions. You have a number of general approaches to attack this problem. This could be classical or quantum Molecular Dynamics, Monte-Carlo methods, etc. For N of some hundreds or so, the required amounts of memory and computing time are too large to handle all the parameters of the system, and some approximations are needed, even for systems of very limited sizes.

On the other hand, if one can suppose from the beginning that the final system will be self-similar, the problem does not arise in the same terms. First, let us discuss the starting hypothesis. It can result from an experimental observation, we know by the experiments that the system will end to the self-similar morphology, and we wish to compute the values of the relevant parameters (e.g. the fractal dimension, the correlation functions, etc.), in order to understand the links existing between these values and the other relevant parameters of the system. Another typical case where one has to state the self-similarity hypothesis is when one is interested by such particular systems (because they have particular behaviors too) and not by the non-fractal ones, since they do not exhibit such scale-invariance. This hypothesis provides an automatic way to sort the fractal systems from the other ones.

Once we have put the self-similarity hypothesis at the very beginning of the intricate and detailed model, one can use the conclusions of the renormalization-group theory. Some parameters are relevant and must be kept without change, while all the others (the irrelevant parameters) can be set to any convenient value. At this point, one has to recognize the relevance quality for any parameter. A simple criterion (which works for most of the parameters) is that any group of parameters which leads to the definition of a characteristic length in the system is made up of irrelevant parameters, as no characteristic length can survive in the self-similar system.

This method leads clearly to a great flexibility in the modelling, allowing a number of variables to be set to any convenient values. We shall see now one example of such a scenario.

MODELLING AGGREGATION OF COLLOIDAL PARTICLES

The fractal morphology of colloidal or aerosol particles has been known for a long time [8]. The basic intuitive idea is that particles dispersed in a fluid move randomly (according to the Brownian motion when the dispersing fluid is the liquid, or the high-density gas), then can stick together by Van der Waals attractive forces. Aggregates can grow sequentially by this way, after successive sticking. As Van der Waals forces are quite strong at this nanometric scale, the aggregates can be assumed to be rigid if not too large.

This scenario is quite simple indeed, but the proper interactions between particles are rather complicated. Schematically (see Fig. 1a) [9] at the contact, the particles are hard objects. A bit farther out, the Van der Waals potential is dominant. Then the possible electrostatic interactions emerge. Other forces can be present (for example indirect forces involving the fluid), and they can be considered and treated in the same way. In principle, the simulations should be performed considering molecular dynamic simulations of all the colloidal particles and all the clusters of particles, interacting through the potential discussed above. This is a hard task, limited presently to a few hundreds of particle, and a short period of time. Instead, one can use the conclusions of the renormalization-group theory to greatly simplify the model.

Since the experimental clusters are known to be fractal, one can concentrate on such a case.

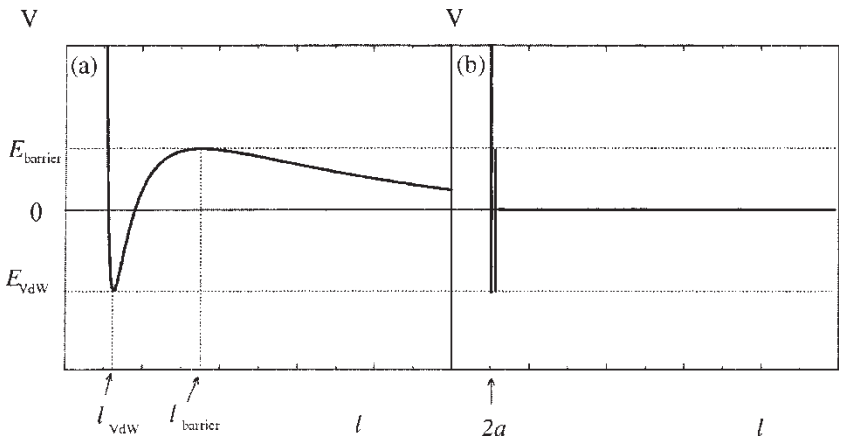

FIGURE 1 (a) Sketch of the 2-particle interaction potential V. It consists essentially of a hard core of diameter $2a$, Van der Waals attractive interaction, and repulsive electrostatic potential. Apart from $2a$, the total potential exhibits two characteristic lengths l_{VdW} and $l_{barrier}$, which correspond respectively to the location of the minimum and the maximum of the potential. (b) The minimal potential energy after removing the two characteristic lengths l_{VdW} and $l_{barrier}$. It is just the hard core—which defines the small length-scale cut-off-, and the minimum and maximum located at contact. The rest of the potential can be set to 0 (see text).

Making then the assumption of spatial self-similarity for the final system, one can remove all the physical parameters leading to a definite characteristic length. We note $2a$, the typical diameter of the primary colloidal particles. This will be the small-length cut-off in the system. We also note m, the mass of one primary particle (this defines the inertia of the particles), and v_o, the typical velocity of the particles and clusters. This defines a characteristic time $\tau = a/v_o$. The first quantity one can remove is the interaction length l_{VdW} corresponding to the minimum of the interaction potential (Fig. 1a). It corresponds to the equilibrium distance between two particles stuck together by Van der Waals forces. Since this must be an irrelevant parameter of the system, one can set its value to any convenient one, for example: $l_{VdW} \to 2a$. This is the same for the maximum of the repulsive electrostatic barrier (when it exists): $l_{barrier} \to 2a$. This corresponds to collapse the whole potential energy onto the diameter hard sphere, as seen in Fig. 1b. Another quantity which can be considerably simplified is the mean free path. As the Brownian motion is fractal itself, it can be replaced by the random walk model, with the mean free path λ equal to either $2a$ (Brownian case, i.e. dense fluid) or ∞ (ballistic regime, i.e. vanishing-density gas). At last, since $\tau^2 E/m$ is the square of a distance, one deduces that the typical energies (namely E_{VdW} and $E_{barrier}$ see Fig. 1a) can be set to 0 or ∞. In fact, the case $E_{VdW} = 0$ is forbidden as it excludes aggregation. We see that just four cases remain to study, and they must correspond to four possible universality classes for the system of colloidal aggregation. Therefore, one expects only four different fractal dimensions to be available for such clusters. Let us discuss briefly the physical meaning of these classes, parametrized by the couple of values $(\lambda; E_{barrier})$.

- $(2a; 0)$ corresponds to Brownian particles. (so, in the dense fluid) with no electrostatic repulsion. This is known experimentally as the rapid colloidal aggregation and leads to a fractal dimension $D_f = 1.8$ [10,11].
- $(2a; \infty)$ corresponds to Brownian particles with the infinite electrostatic repulsive barrier to pass through, in order to aggregate. Of course, this can only be considered as a limit case, where the barrier is very high. The clusters or the particles have to hit many times before getting a finite probability to stick together (when the relative kinetic energy is larger than the barrier). This corresponds in fact to the vanishing probability of irreversible sticking, and is known experimentally as the slow colloidal aggregation regime. The corresponding fractal dimension is $D_f = 2.0$ [10,11].
- The two other cases, with $\lambda = \infty$, correspond to the similar respective experiments in the rarefied fluid (aerosol aggregation), and the fractal dimensions are $D_f = 1.9$ and 2.1, respectively [10,11].

This minimal model with the four possible sets of relevant parameters is known as "Cluster–Cluster Aggregation model" [12]. It is briefly described in the Appendix, and a typical simulated aggregate is shown in Fig. 2. One should remark that the numerical model is free from any numerical parameter, even if it is able to give easily accurate values of relevant physical quantities (such as the fractal dimension).

One can note that this minimal model was first introduced as a tentative approximated model to understand the fractal structure of the colloidal and aerosol aggregates [13,14]. The present approach allows us to explain the amazing agreement of the numerical and experimental data, as well as the very

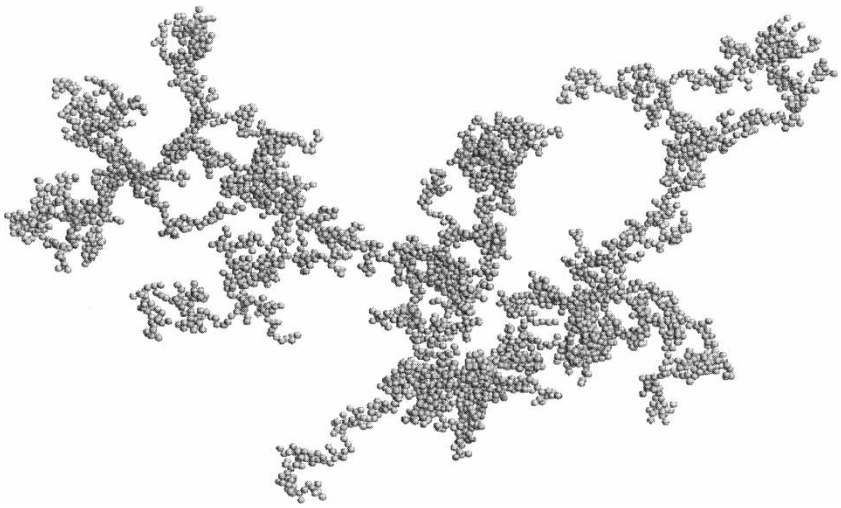

FIGURE 2 Image of a cluster obtained numerically by the minimal model described in the "Appendix" Section, $N = 4086$ particles. The fractal dimension $D_f = 1.8$.

limited number of possible fractal dimensions according to the physical conditions. Moreover, it offers a general explicit scenario for the other (self-organized) critical systems.

CONCLUSION

From the original molecular dynamics simulations, with its complete set of continuous parameters, the only hypothesis of spatial self-similarity of the final system allowed us to consider Monte-Carlo simulations with a minimal number of relevant parameters taking only simple values. The rest, in particular the interaction potentials, is made quite simple by the procedure (e.g. just potentials at contact), and this permits large-scale simulations, with system sizes increased by several order of magnitudes in comparison with molecular dynamics. In addition, the values of the fractal dimensions, of the correlation functions, of all the physical quantities linked to the self-similar structure of the final system, are exactly the same as for the initial molecular dynamics (if this was possible for such large systems), or to the experiments. For the critical systems, this procedure is not an approximation but a mapping scheme, able to replace a system that is quite difficult to handle by a much simpler equivalent system. For the systems which are not critical but close to the critical point, a similar procedure (following the renormalization-group theory) can be used, but now just as an approximation scheme. For such systems, irrelevant parameters must then be seen as parameters which can be safely approximated. Unlike the critical case, and even if the method can be still very efficient, one cannot avoid the difficult task of the control of the approximations in the non-critical systems.

APPENDIX

Here, we should like to describe briefly the Cluster–Cluster Aggregation Model in its simplest version, for the case $(\lambda; E_{\text{barrier}}) = (2a; 0)$. The changes needed for the other three cases described in the text are straightforward.

Let us consider a cubic box size L, with periodic boundary conditions. Inside the box we put randomly N spherical particles of diameter $2a = 1$ (this will define the unit of length).

- The particles and clusters will diffuse according to random walk, with the mean free path 1, and velocity $v_0 = 1$ (this will define the unit of time). The diffusion is performed through Monte-Carlo method: one particle or cluster, say C_o, is chosen randomly with equal probability. C_o is moved following a straight trajectory of length 1, in a random direction.
- If during the movement of C_o there is an overlap with another particle, say P, then the movement is performed only until close contact between C_o and P is realized. The new cluster built in this way by the geometric union of C_o and of P (or the cluster to which P belongs) will be rigid and never break up.
- The process continues until there is just one cluster of N particles in the box.

References

[1] Havlin, S. and Bunde, A. eds (1995) *Fractals in Science* (Springer-Verlag, New York).
[2] Duran, J. (2000) *Sands, Powders and Grains: An Introduction to the Physics of Granular Materials* (Springer-Verlag, New York).

[3] Mandelbrot, B.B. (1982) *The Fractal Geometry of Nature* (W.H. Freeman and Co., New York).
[4] Suzuki, M. (1982) "Phase transition and fractals", *Prog. Theor. Phys.* **69**, 65.
[5] Jensen, H.J. (1998) *Self-Organized Criticality*, Cambridge Lecture Notes in Physics (Cambridge University Press).
[6] Weitz, D.A., Hunag, J.S., Lin, M.Y. and Sung, J. (1985) "Limits of the fractal dimension for irreversible kinetic aggregation of gold colloids", *Phys. Rev. Lett.* **54**, 1416.
[7] Wilson, K.G. and Kogut, J. (1974) "The Renormalization-Group and the ε-expansion", *Phys. Rep. C* **12**, 75.
[8] Forrest, S.R. and Witten, T.A., Jr. (1979) "Long-range correlations in smoke-particle aggregates", *J. Phys. A* **12**, L109.
[9] Isrealachvili, J. (1991) *Intermolecular and Surface Forces* (Academic Press, London).
[10] Botet, R. and Jullien, R. (1990) "Fractal aggregates of particles", *Phase Transition* **691**, 24–26.
[11] Meakin, P. (1998) *Fractals, Scaling and Growth far From Equilibrium* (Cambridge University Press).
[12] Jullien, R. and Botet, R. (1987) *Aggregation and Fractal Aggregates* (World Scientific, Singapore).
[13] Meakin, P. (1983) "Formation of fractal clusters and network by irreversible diffusion-limited aggregation", *Phys. Rev. Lett.* **51**, 1119.
[14] Kolb, M., Botet, R. and Jullien, R. (1983) "Scaling of kinetically growing clusters", *Phys. Rev. Lett.* **51**, 1123.