Non-equilibrium cluster growth: where did it all begin

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1. Introduction

In this paper, Forrest and Witten reported on an experimental study of metal particle aggregates formed when a metal vapor produced by heating a plated filament was quench condensed. Metal particles of average radius 40Å were streamed outward and accumulated in a thin spherical shell (a "puff ball") of radius ~ 1 cm where the particles coalesced. The resulting particle aggregates moved down under under gravity to an electron microscope slide where they were subsequently examined.

The aggregates were found to have fractal shapes, which visually call for immediate analogies and possible interpretations in terms of the percolation clusters, self-avoiding random walks, dendrites, etc. The black and white images obtained by the electron microscope were digitized and analysed by measuring their box-counting (essentially Hausdorff) dimension, which was found to be in the range 1.5 to 1.6. Auto-correlations of the aggregate density were measured attributing density $\rho = 1$ to the black and $\rho = 0$ to the white colours on the images. It was reported that, similarly to other previously known extended systems with critical behaviour, the aggregates exhibited long correlations with a power-law behaviour

$$\langle \rho(r)\rho(0)\rangle - \langle \rho(0)\rangle^2 \sim r^{-A}$$

with fractional exponents A which were found to be ≈ 0.32. Both the correlation measurement technique and the box-counting method where calibrated using test images with known properties, the Koch curve and a plot with random position of dots, and the error bars were estimated.

The authors exposed striking properties suggesting that the considered system is governed by a critical process, and offered a candidate for such a process: a percolation mechanism acting in the "puff ball". On the other hand, they were cautious enough by leaving a possibility for interpretations which are unrelated to the critical phenomena.

It is clear that this is a pioneering paper which has had a huge impact (717 citations in the Google Scholar database) and served as an inspiration for the subsequent development of the theoretical, computational and experimental works in non-equilibrium statistical mechanics and critical phenomena. It helped shaping new areas, such as the diffusion-limited aggregation, kinetic aggregation, dendrites, scale-invariant cluster percolation. The experiment was very simple, and yet very creative, yielding clear and beautiful thought-provoking results.

As a matter of fact, a powerful impact of the J. Phys A paper by Forrest and Witten was observed almost instantly after its publication. Inspired by this paper and building on its findings, in 1981 Witten and Sander published a PRL "Diffusion-Limited Aggregation, a Kinetic Critical Phenomenon" in which they put forward a new discrete dendric growth model and presented numerical simulations, which produced brownian trees looking similar to those observed in the experiments of Forrest and Witten and having a similar fractal dimension [1]. This started the era of the diffusion-limited aggregation (DLA), which is presently one of the important research directions in the

areas of mathematical physics and statistical mechanics. The research on DLA has since been experiencing a vigorous growth with large number of papers published every year. It is clear that the experimental discoveries of the paper by Forrest and Witten were of a trailblazing nature for this field, and its impact is not surprising.

[1] Witten, T. A. and Sander, L. M., Diffusion-Limited Aggregation, a Kinetic Critical Phenomenon, Phys. Rev. Lett., vol 47, issue 19, pages 1400–1403, (1981)