

CECAM-Workshop on Kinetic Models for Cluster Formation: Orsay, September 17–28, 1984

R. Jullien,¹ M. Kolb,¹ H. Herrmann,² and J. Vannimenus³

We present the abstracts of the talks given at the workshop on kinetic models for cluster formation held at Orsay.

The field of cluster growth and aggregation phenomena has been rapidly expanding in the last few years. Since the introduction of the first growth model for diffusion-limited aggregation (in which Brownian particles stick one by one on a single growing cluster), many other models have been introduced and there have been systematic numerical investigations of the fractal properties of the resulting clusters. In parallel a growing body of experiments has been investigated and compared with theory. Still the field is a young one and rather phenomenological. The first thrust went into devising as many models as there are different experimental situations, with the result that by now large portions of the real axis are paved with fractal dimensions (when allowing for error bars). A good overview of the status of the field, before the workshop, can be found in the proceedings of the conference on kinetics of aggregation and gelation, held in Athens, Georgia in April 1984 (these proceedings have been edited by F. Family and D. L. Landau and published by North-Holland). In the meantime much progress was made which motivated us to organize a workshop on the subject.

This workshop was held in Orsay from the 17th to the 28th of September. It was sponsored by the CECAM (Centre Européen de Calcul Atomique et Moléculaire, Bât. 506, Orsay, France). The aim of this meeting was to give ample time and opportunity for consolidating the different

¹ Physique des Solides, Bât. 510, Université Paris-Sud, 91405 Orsay, France.

² Physique Théorique, CEN Saclay, 91191 Gif sur Yvette, France.

³ Groupe de Physique des Solides de l'E. N.S., 24, rue Lhomond, 75005 Paris, France

approaches, experiments, and theories, and to try to develop a more systematic way to describe aggregation phenomena.

The talks, usually in the morning, were followed by round tables and informal discussions in the afternoons. The topics covered included direct experimental observations of growth processes on a microscopic as well as on a macroscopic scale and addressed the question "how can one best extract the fractal properties?" There were theoretical considerations on the role of the surface of growing aggregates, the influence of fluctuations and randomness, as well as general self-similarity and universality properties. While the bulk of the models proposed still rely on numerical simulations, there are a few exact results in some special cases and some encouraging attempts at field theoretic formulations. We now summarize the talks of the workshop where the names in parentheses make reference to the speaker. The corresponding abstracts are given later on.

Many interesting experiments were presented at this workshop. Some of them can be accounted for by the particle-cluster aggregation model, as originally proposed. They include dielectric breakdown (L. Pietronero) and copper electrodeposit (R. Brady). The mechanism of cluster-cluster aggregation, either kinetically or chemically controlled, has been realized experimentally, and both static and dynamic properties were determined (D. Weitz, Z. Djordjevic). For various macroscopic model systems, the geometrical structure of growing clusters were determined: aggregation of latex spheres (P. Richetti) wax balls (C. Allain, R. Blanc), filtration experiments (D. Houi), and flow in a viscous medium (J. Nittmann). The question of how to analyze the experimental data was addressed by studying the optical Fourier transform of fractals (M. Cloître).

On the theoretical side, let us first mention the progress made in understanding the general properties of fractal structure in growth. In addition to the fractal dimension several other exponents have been defined and useful relations between them could be established (H. E. Stanley). The influence of randomness (T. Witten) and the nature of the interface (L. Sander, B. Sapoval) were thoroughly discussed. Questions of universality and self-similarity have been investigated by the renormalization group (M. Kolb). Elastic (H. Herrmann) and electrical (P. Rammal) properties of fractal structures were also considered. Exact results for aggregation are limited to trees, where the concepts can be illustrated quite clearly (V. Hakim, J. Vannimenus).

Much theoretical effort has been made to refine existing models and to evaluate more detailed properties. The so-called ballistic aggregation models have been investigated numerically and qualitatively (L. Sander). Kinetic gelation has been quantitatively compared with equilibrium gelation (D. Landau). Size distribution of clusters in cluster-cluster

aggregation has been investigated numerically in great detail (P. Meakin). Moreover, the cluster-cluster aggregation model has been extended to the case of two compounds (Z. Djordjevic, P. Meakin) and to the limit of a vanishing sticking probability (R. Ball, R. Jullien) with some successful agreement with experiments. The different role played by particle-cluster and cluster-cluster aggregations has been elucidated (R. Ball, R. Botet). A number of variants of kinetically growing self-avoiding walks have been studied, mainly to test which aspects are relevant for changing the fractality (A. Coniglio, J. Lyklema) and to try to make a field theory for growth processes (L. Peliti). It was also shown that the models used for growth processes have other interesting applications, especially in epidemiology and social behavior (P. Grassberger).

The kinetic approach to aggregation provides a useful alternative for studying growth problems. Based on the Smoluchowski equation, this analytical approach helps to classify the different scaling regimes and provides some exact answers for specific growth models (P. van Dongen, M. Ernst, F. Leyvraz, E. Hendriks).

Below, we give a list of the participants with their addresses and the abstracts of the talks (alphabetically). References for the talks are given at the end.

LIST OF PARTICIPANTS

R. C. BALL	Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, England
D. BIDEAU	GEPM, Université de Rennes, 35032 Rennes Cédex, France
R. BOTET	Physique des Solides, Bât. 510, Université Paris XI, Orsay, France
R. M. BRADY	Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, England
C. CAMOIN	Département de Physique des Systèmes, Université de Provence, Marseille, France
X. CAMPI	I.P.N., Bât. 100, Université Paris XI, 91505 Orsay, France
J. N. CHAZALVIEL	École Polytechnique, Palaiseau, France
M. CLOITRE	École de Physique et Chimie, Lab. d'Hydrodynamique, 10, rue Vauquelin, 75005 Paris, France

- A. CONIGLIO Istituto di Fisica Teorica, Mostra d'Oltremare,
Pad. 19, 80125 Napoli, Italy
- H. CORNILLE DPMT, CEA Saclay, 91191 Gif-sur-Yvette,
France
- B. DERRIDA SPT, CEA Saclay, 91191 Gif-sur-Yvette,
France
- G. DIETLER ETH Zürich, CH-8093 Zürich, Switzerland
- Z. DJORDJEVIC Institut "Boris Kidric" Vinca, POB 522, 11001
Belgrade, Yugoslavia
- P. G. J. van DONGEN Inst. for Theor. Physics, Univ. Utrecht, Prin-
cetonplein 5, 3508 TA Utrecht, The
Netherlands
- M. ERNST Inst. for Theor. Physics, Univ. Utrecht, Prin-
cetonplein 5, 3508 TA Utrecht, The
Netherlands
- A. P. GAST École de Physique et Chimie industrielle, 10,
rue Vauquelin, 75005 Paris, France
- J. F. GOUYET École Polytechnique, Palaiseau, France
- P. GRASSBERGER Physics Dept., Univ. Wuppertal, D-56 Wup-
pertal 1, Federal Republic of Germany
- V. HAKIM ITP Santa Barbara and LPJHE, Orsay,
France
- R. HENDRIKS Institut für Theoretische Physik, Zülpicher-
strasse 77, 5000 Köln 41, Federal
Republic of Germany
- H. J. HERRMANN Service de Physique Théorique, CEN Saclay,
91191 Gif-sur-Yvette, France
- D. HOUÏ Institut de Mécanique des Fluides, 2, rue C.
Comichel, 31071 Toulouse Cédex, France
- R. JULLIEN Physique des Solides, Bât. 510, Université
Paris XI, Orsay, France
- M. KOLB Physique des Solides, Bât. 510, Université
Paris XU, Orsay, France
- Y. KURAMOTO RIFP Kyoto University, Kyoto, Japan
- D. P. LANDAU Dept. of Physics and Astronomy, Univ. of
Georgia, Athens, Georgia 30602
- F. A. LEYVRAZ Center for Polymer Studies, Boston Univ.,
Boston, Massachusetts 02215
- J. W. LYKLEMA IFF-KFA, D 5170 Jülich, Federal Republic of
Germany
- P. MANNEVILLE DPh-G/PSRM, CEA Saclay, 91191 Gif-sur-
Yvette, France

- H. MARTIN École Normale Supérieure, Groupe de Physique des Solides 24, rue Lhomond, 75231 Paris Cédex 05, France
- P. MEAKIN BLOG 356 Rm 251 CR & DD Dept. Experimental Station E. I. Du Pont de Nemours & Co, Wilmington, Delaware 19898
- J. NITTMANN Dowell-Schlumberger, Z. I. Molina la Chasatte, 42003 St-Etienne, France
- N. OSTROVSKI Lab. de Physique de la Matière Condensée, Univ. de Nice, Parc Valrose, 06034 Nice Cédex, France
- L. PELITI Dipartimento di Fisica, Università "La Sapienza," Piazzale Aldo Moro 2, I-00185, Roma, Italy
- L. PIETRONERO Solid State Physics Laboratory, University of Groningen, 9718 EP, Groningen, The Netherlands
- R. RAMMAL CNRS-CRTBT, 166X, 38042 Grenoble Décex, France
- P. RICHETTI CRR, Domaine Universitaire de Bordeaux I, 33400 Talence, France
- M. ROSSO École Polytechnique, Palaiseau, France
- B. ROULET M. P. S., Univ. Paris VII, place Jussieu, 75231 Paris Cédex 05, France
- M. SALEM SGT, CEN Saclay, 91191 Gif-sur-Yvette, France
- L. SANDER Physics Dept., Univ. of Michigan, Ann. Arbor, Michigan 48109
- B. SAPOVAL École Polytechnique, Palaiseau, France
- M. SORNETTE Lab. de Physique de la Matière Condensée, Université de Nice, Parc Valrose, 06034 Nice Cédex, France
- H. STANLEY Center for Polymer Studies, Boston Univ., Boston, Massachusetts 02215
- J. VANNIMENUS École Normale Supérieure, Physique des Solides, 24, rue Lhomond, 75231 Paris Cédex 05, France
- D. WEITZ Exxon Research and Engineering, Rt 22E Annandale, New Jersey 08801
- T. WITTEN Exxon Research and Development, Annandale, New Jersey 08801

ABSTRACT OF THE TALKS

These are given in alphabetical order of the speakers. The speakers are underlined.

C. ALLAIN, R. BLANC, C. CAMOIN, and E. GUYON, *Macroscopic aggregation experiments*: The formation of clusters in systems of particles is a very common process involved for instance in the flocculation of colloids, coagulation of aerosols or in radical chemical reactions. In this report, we present two sets of analog simulations of two-dimensional clusters which take into account physical processes not easily described by numerical works.

In a first experiment,⁽¹⁾ we have studied the growth nine-dimensional clusters of 1-mm diameter spheres floating on a surface of water. Because of the attractive capillary forces between these spheres, when two balls collide, they stick together permanently. At the beginning of the simulation, the balls are distributed randomly on the surface. During the experiment, their motion is due to random hydrodynamic movements of the surface, the Brownian effects being negligible. The existence of a critical surface concentration for the appearance of a cluster extending from one edge of the container to the opposite one has been proved. For this concentration (0.195), the clusters exhibit self-similar properties with a fractal dimension equal to 1.64. It is worth noting that this value is larger those obtained in numerical simulations. This can be related to intracluster aggregation processes and to hydrodynamic interactions which have not been taken into account in the numerical simulations.

In a second experiment,⁽²⁾ the suspension of spheres is submitted to a shear flow. There is a competition between hydrodynamic forces which bring the particles together and separate them and the capillary forces which tend to aggregate them. Different clustering behaviors have been observed depending on the relative importance of these two forces. The amplitude of the capillary forces can be varied by changing the thickness d of a layer of oil having the same density as the spheres. If d is equal to the diameter of the spheres, no attraction is observed and hydrodynamic clusters of finite lifetime are formed.⁽³⁾ In the limit of zero shear rate and low capillary forces, results are identical to those of the first experiment. In intermediate situations, compact clusters are formed.

R. BOTET, R. JULLIEN, M. KOLB, *Connection between particle-cluster and cluster-cluster aggregation*: An extension of the hierarchical model of clustering of clusters⁽⁴⁾ is introduced to allow direct numerical simulations of kinetic aggregation with any sort of kinetic coefficients K_{ij} . Studying the example $K_{ij} = (ij)^\omega$, we show⁽⁵⁾ that the DLA model is recovered when there is gelation ($\omega > 1/2$). When there is no

gelation ($\omega < 1/2$), we find numerically clusters with the typical fractal dimensionality of the clustering of clusters process. Nevertheless, the case where $0 < \omega < 1/2$ is not quite clear since the analytical derivation of the mean field value of the fractal dimensionality of the clusters above the upper critical dimension^(6,7) yields to continuously varying exponent.⁽⁸⁾ The use of other sorts of parameters to distinguish between the different classes of models is pointed out.

R. M. BRADY and R. C. BALL, *Fractal growth of copper electrodeposits*: Black dendritic growths of copper were electrodeposited onto an initially pointlike cathode. The electrolytic solution contained Cu^{2+} ions, an excess of Na^+ and SO_4^{2-} ions to screen out any electric fields, and an inert viscosity enhancer to prevent stirring of the solution. It was found that the reaction rate was limited by diffusion of copper ions.

As the deposit grew, its mass M could be monitored by measuring the total charge transferred, and its average radius R could be found since it was proportional to the instantaneous current. Fractal behavior was observed, $M \propto R^D$, with $D = 2.43 \pm 0.03$, in good agreement with computer simulations of diffusion-limited aggregation.

The potential for further work based on these experiments will be discussed.

C. ALLAIN, M. CLOITRE, *Optical Fourier transforms of fractals*: We describe how to realize Fourier's transforms of fractals, in order to characterize clusters which are obtained in a great number of aggregation processes. OFT is based on the capacity of a convergent lens to realize a Fourier's transform. This analogic technique is extremely convenient compared to other methods which are usually employed to study fractals (for instance, correlations calculi, applications of the mass-radius relation).

Here, we restrict ourselves to the case of two-dimensional deterministic fractals, such as Cantor's sets and Serpinski's carpets. The fractal objects that we are studying are first calculated on a microcomputer and drawn by means of a graphics plotter. Then, we take photographs of these drawings, using 1 24 × 36-mm-high resolution film. Finally, the so-obtained objects (OB) are placed in an optical Fourier's transform arrangement and we study the repartition of their spatial frequencies in the reciprocal Fourier space.

Two sorts of information are available from these Fourier's transforms, since we observe (1) intense quasiperiodic spots whose repartition in the Fourier place exhibits the same symmetry elements as OB, and (2) a continuous, "fractal," frequency spectrum which is due to the fractal nature of OB. Moreover, in the special cases of Cantor's sets and Serpinski, carpets, the Fourier transforms can be calculated analytically and compared to the optically obtained spectra. In view of this, we show how to determine

directly on the spatial spectrum of OB the Hausdorff's dimension and the smaller and larger scales of OB.

Finally, from this study, we can discuss precisely the application of OFT as an image processing system, to analyze clusters, aggregates, which are observed in a lot of experimental investigations.

A. CONIGLIO, *Kinetic walk models and their relevance to polymers and gels*: Recently introduced kinetic walk models are reviewed. Their relevance is shown to describe (i) the irreversible growth of a linear polymer, (ii) the coil globule transition which occurs at the θ temperature, and (iii) the structure of a gel.

G. DIETLER, *Experiments on blood coagulation*: The aggregation of fibrin induced by the enzyme thrombin was investigated by combining static and dynamic light scattering with measurements of the release of the fibrinopeptides A.

We present experiments which show that the reaction coordinate is the number of reactive binding sites per monomer initially in solution. Since the cluster size distribution is in equilibrium at every time, the aggregation is controlled by the action of the enzyme thrombin and the kinetic of aggregation reduces to the kinetic of the enzyme action.

These results are valid only for concentrated solutions (fibrinogen concentration ~ 2 mg/ml), where the mean distance between two monomers is comparable to the dimension of the monomers and the monomers do not need to diffuse in order to meet another monomer to form a bond.

Z. B. DJORDJEVIC, *Scaling picture of polymerization kinetics*: The kinetics of polymerization in dilute mixtures is monitored both by measurements of the intensity of the light scattered in the forward direction, which provides the information on the second moment of the cluster size distribution, as well as by the infrared spectroscopy, which gives the mean number of chemical bonds. It is found that both the intensity of the scattered light and the intensity of the monitored absorption lines grow as power laws of time. The former with an exponent y_2 equal to 2.6 ± 0.1 and the later with $y_1 \sim 0.8 \pm 0.1$. These results can be explained by the scaling picture of the polymerization kinetics which predicts that the mean number of clusters with b - chemical bonds scales as $n_b(t) = b^{-\theta} f(b/t^z)$ where t is the reaction time and θ and z are two exponents related to measured exponents y_2 and y_1 . The corresponding values are found to be $\theta \sim 1.6 \pm 0.1$ and $z \sim 1.9 \pm 0.2$. Also, these exponents are expressed as functions of the fractal dimension of the polymers D and the diffusion constant exponent α . The values of the latter are deduced to be $D = 1.84 \pm 0.1$ and $\alpha = -0.47 \pm 0.1$.

Z. B. DJORDJEVIC and P. MEAKIN, *Gelation in $A_2 + B_f$ systems*: Kinetics of aggregation and gelation as well as structure of

polymers created in the mixtures of two types of polyfunctional monomers are investigated. We have developed a mean field kinetic theory for these processes and performed computer simulations in both two and three dimensions. It is discovered that gelation occurs only for the certain, limited, range of the values of the relative concentration of two components. Outside, of this range polymerization results only in finite clusters. Apparent fractal dimension of the aggregates appears to be dependent on the composition of the chemical mixture. The same is true for the exponent z characterizing time evolution of the distribution function $n(t) = S^{-\theta} f(s|t^z)$.

G. J. van DONGEN and H. ERNST, *Asymptotic solutions of the coagulation equation*: The cluster size distribution $c_k(t)$ in a system of reacting clusters, $A_i + A_j \rightarrow A_{i+j}$, is studied using Smoluchowski's coagulation equation, where $K(i, j)$ are the coagulation coefficients. We study the class of homogeneous kernels $K(i, j)$, characterized by the exponents λ, μ with $2\omega = \lambda + \mu$:

$$K(si, sj) = s^{2\omega} K(i, j) \tag{1}$$

$$K(i, j) \simeq j^{\lambda, \mu} \quad (j \rightarrow \infty, i \text{ fixed})$$

The reactivity of *large* clusters should not increase faster than their size; thus $\omega \leq 1, \lambda \leq 1$ (no restrictions on μ).

The *short-time behavior* ($t \downarrow 0$) of $c_k(t)$ for initially monodisperse systems is described by $c_k(t) \simeq a_k t^{k-1}$, where the positive numbers a_k satisfy $a_1 = 1$ and⁽⁹⁾

$$(k-1)a_k = (1/2) \sum K(i, j) a_i a_j \quad (i+j=k) \tag{2}$$

The ansatz $a_k \simeq AK^{-\theta} R^{-k}$ ($k \rightarrow \infty$) yields self-consistently $\theta = 2\omega$ for all $\lambda < 1$.⁽¹⁰⁾ On the *singular line* $\lambda = 1$ the θ exponent (which is not longer uniquely determined by λ and μ) can be solved from a transcendental equation involving $K(i, j)$ explicitly. The solution may assume any value $\theta \in (\mu, \infty)$, e.g., $\theta = (2 + \alpha + \mu)/2$ for $K(i, j) = (i^\mu j^\alpha + j^\mu i^\alpha)(i+j)^{1+\alpha}$ with $\mu < \alpha$. Both A (and even its sign) as well as R are only known for a few solvable models, e.g., $K(i, j) = (ij)^\mu (i+j)^{1-\mu}$ yields $a_k \simeq k^{-\theta} e^{k/\sqrt{2\pi}}$ with $\theta = \mu + 3/2$. The θ exponent describes the size distribution $c_k \simeq k^{-\theta} a^k$ for "lattice animals" or branched polymers a finite time before t_c [$t_c < \infty$ in gelling systems ($\omega > 1/2$), and $t_c = \infty$ in nongelling systems ($\omega \leq 1/2$)]. A possible method to obtain the *long-time* ($t \rightarrow \infty$) or *critical* ($t \rightarrow t_c$) behavior of c_k is to make the ansatz that $v_k(t) = c_k(t)/c_1(t)$ approaches a positive number b_k as $t \rightarrow \infty$, yielding⁽¹¹⁾

$$b_k \sum_{j=1}^{\infty} [K(k, j) - K(1, j)] b_j = \frac{1}{2} \sum_{i+j=k} K(i, j) b_i b_j \tag{3}$$

For gelling systems Eq. (3) applies also to the special postgel ($t > t_c$) solution, $c_k(t) = b_k/[1 + \beta(t - t_c)]$.⁽¹²⁾ Simple kernels are $(ij)^\lambda$ and $i^\lambda + j^\lambda$. We look for solutions of the form $b_k \simeq Bk^{-\tau}\beta$ ($k \rightarrow \infty$), and self-consistency arguments yield for homogeneous kernels with $\mu > 0$: $\tau = 1 + 2\omega$ ($\omega < 1$)⁽¹³⁾ and $\tau = \omega + 3/2$ ($1/2 < \omega < 1$). On the *singular line* $\mu = 0$, the τ value is no longer uniquely determined by $\omega = \lambda/2$. For ($0 < \lambda < 1$, $\mu = 0$) τ can be solved from a transversal equation involving $K(i, j)$. Possible solutions lie in the interval $(1, 1 + \lambda)$.

An *alternative method* for obtaining asymptotic properties of $c_k(t)$ is via similarity solutions of Smoluchowski's equation, the so-called *self-preserving spectra*. Here we consider the region $\lambda < 1$. For nongelling ($\omega = 1/2$) systems $c_k(t) = g^2\phi(kg)$ with $x = kg(t)$ kept fixed as $k, t \rightarrow \infty$, where $g(t) \sim t^{1/\sigma}$ with $\sigma = 2\omega - 1$. For gelling ($\omega > 1/2$) systems, $c_k(t) \simeq g^\tau\phi(kg)$ with $x = kg(t)$ kept fixed $k \rightarrow \infty$ and $t \uparrow t_c$, where $g(t) \sim (t_c - t)^{1/\sigma}$ with $\sigma = \omega - 1/2$. The behavior of the *scaling function* as $x \downarrow 0$ is $\phi(x) \sim x^{-2\omega} \exp(-x^\mu)$ if $\mu < 0$, and $\phi(x) \sim x^{-\tau}$ with $\tau = 1 + 2\omega$ if $\mu > 0$ and $\omega < 1/2$; $\tau = \omega + 3/2$ if $\mu > 0$ and $1/2 < \omega < 1$. In the (λ, μ) region with $\lambda < 1$ one has $\phi(x) \sim x^{-2\omega} \exp(-x)$ as $x \rightarrow \infty$.

One could try, e.g., for nongelling systems, to calculate $c_k(t)$ as $t \rightarrow \infty$ at fixed k , or $k \rightarrow \infty$ at fixed t from the similarity form $c_k(t) = g^2(t)\phi(kg(t))$. This yields $\theta = 2\omega$ for all $\lambda < 1$, in agreement with the recursion (2). The long-time behavior of $c_k(t)$ at fixed k for nongelling systems ($\omega < 1/2$) corresponds to the small- x behavior of $\phi(x)$ and yields $c_k(t) \sim t^{-\gamma}k$ with $\gamma = +1$ for ($\mu > 0$, $\lambda < 1$), and $\gamma = (2 - \tau)/(1 - \lambda)$ for ($\mu = 0$, $0 < \lambda < 1$). In the latter case ($\mu = 0$) the γ and τ exponent depend on the detailed form of $K(i, j)$.

It appears also possible to construct fragmentation kernels $F(i, j)$, appropriate to $K(i, j)$.⁽¹⁴⁾ Fragmentation processes do not affect the short-time behavior; for $\omega > 1/2$ and sufficiently large fragmentation, gelation is suppressed; for $\omega < 1/2$ fragmentation processes prevent $c_k(t)$ from reaching its self-preserving form.

P. GRASSBERGER, *Cluster growth in social phenomena and epidemics*: I shall review the relation between two models of epidemics and directed (resp. undirected) percolation. Also, I shall review the connection between coalescing random walks and public opinion formation. After that, I shall discuss a field theoretic formulation which should, e.g., enable one to compute the ε expansion of the "intrinsic spreading dimension" in percolation.

Finally, I shall discuss modifications suggested in the literature on epidemics, and ask how they change critical behavior. Some lead to various interacting Levy flights.

V. HAKIM, **B. NICKEL**, **J. VANNIMENUS**, *Diffusion limited*

aggregation on the Cayley tree: We study the diffusion-controlled process of cluster growth, introduced by Witten and Sander, on a Cayley tree. We show that it is then equivalent to the Eden model where growth occurs at any boundary site with equal probability. The mean level number and the square gyration radius of an N -particle aggregate both increase as $[K/(K-1)] \ln N$ on a tree of branching ratio K . The case of biased diffusion is studied numerically: an attractive bias does not change the logarithmic behavior of the size, but a repulsive bias leads to a different behavior, presumably with a mean level number of order N .

E. M. HENDRIKS, *Cluster size distributions in equilibrium:* Equilibrium size distributions for finite system containing M monomers in a volume V undergoing coagulation (polymerization)/fragmentation, are determined for coagulation rate K_{ij} and fragmentation rate F_{ij} , corresponding to the process $A_i + A_j \rightleftharpoons A_{i+j}$ (here A_k denotes a cluster containing k monomeric units). It is shown that microscopic detailed balance implies $F_{ij}/K_{ij} = \lambda a_i a_j / a_{i+j}$, where λ and e_k are arbitrary. The mean and most probable size distribution $c_k = \lambda \rho^{-1} a_k e^{-\mu k}$ coincide in the thermodynamic limit $M, V \rightarrow \infty$, $\rho = M/V$ fixed. If $a_k \sim e^{\mu c} k^{-\delta}$ ($k \rightarrow \infty$), $2 < \delta < 3$, then the theory predicts a (sol-gel) phase transition at a well-defined value of $q = \lambda \rho^{-1}$, with critical exponents $\tau = \delta$ and $\sigma = \delta - 2$. The gel fraction exponent β assumes its classical value unity, probably due to the neglect of spatial fluctuations. Finally it is indicated how the theory can in principle be extended to account for isomerism and cyclization.

H. J. HERRMANN, *Elasticity of random media:* There is not one single theory for the elasticity of a random medium. The recent understanding in this complex field are reported. Particular emphasis is made on the geometrical character of the elastical part of a network. The backbone and the elastic backbone of percolation cluster are investigated numerically. Their unusual behavior explains the special features of some models for elasticity. The growth model of kinetic gelation has a considerably larger fractal dimension of the backbone than percolation. Thus one expects different elastic and other dynamic exponents for kinetic gelation than those usually calculated for percolation.

D. HOUJ and M. LENORMAND, *Particle deposit during filtration:* Cake filtration is the continuous accumulation of solid particles at the surface of a filter medium.

The growth and properties of this deposition are studied through experimental and computer simulations with specific rules which take into account physical mechanisms.

The superposition of a ballistic displacement, due to the fluid flow and a diffusive motion (Brownian motion) is described by a Peclet number.

Simulations have been carried out to investigate the crossover of r.m.s.

thickness exponent ε between ballistic ($\varepsilon=1$) and diffusive ($\varepsilon=1.3$) deposition.

R. JULLIEN and M. KOLB, *Chemically limited cluster-cluster aggregation*: The chemically limited cluster-cluster aggregation model is defined as a limiting model of diffusion-limited aggregation when the sticking probability becomes infinitesimally small. This model can also be viewed as a diffusive model in which the fractal dimension s_w of the relative trajectory of the clusters tends to zero. Numerical investigations have been done in $d=2, 3, 4$ on a hierarchical version of the model.⁽¹⁵⁾ The fractal dimension of the clusters, D , as well as the exponent showing how the number of active sites N_A increases with the total number of particles N in a cluster ($N_A \sim N^\delta$) have been determined. Then, the crossover between Brownian cluster-cluster aggregation and chemically limited aggregation, when varying the sticking probability has been numerically investigated in a box.⁽¹⁶⁾ In three dimensions the fractal dimension increases from $D \sim 1.78$ (diffusive) to $D \sim 2.00$ (chemically limited) in good agreement with the experiments of Weitz.

M. KOLB, *Renormalization group for aggregation*: A general renormalization group transformation (RG) for the static as well as the kinetic properties of aggregates is formulated and applied to several growth models: particle aggregation, cluster aggregation, and growth percolation. The transformation is implemented using the Monte Carlo renormalization in real space and the RG flow is monitored utilizing several short-range static and dynamic correlations. Crossover behavior can be observed and critical exponents can be evaluated this way.

D. P. LANDAU, *Growth models for addition polymerization*: Growth models for addition polymerization which explicitly include kinetics have recently been proposed and explored. Extensive computer simulation data now exist for lattice models in two and three dimensions for which polymerization occurs primarily through radical initiated growth. Results for bulk properties have been obtained in the vicinity of the gel-sol transition for a wide range of lattice sizes and initiator site concentrations. We shall review recent work on these models and compare and contrast the results with those for random percolation and Flory-Stockmayer theory.

F. LEYVRAZ, *Rate equation and aggregation process*: An aggregation process can be described by the scheme



where A_j is an aggregate of size j and R_{jk} are suitably chosen rate constants. This leads to rate equations for the concentrations c_j of A_j as a function of time. It is found that such equations lead to a scaling behavior

of the type observed in actual computer simulations. Some variations on that model—involving, e.g., time-dependent reaction rates—can also be discussed. For a wide class of such models the different critical exponents involved can be evaluated exactly.

J. W. LYKLEMA and K. KREMER, *Irreversible self-avoiding walks*: We discuss two new self-avoiding walks, which possess the property of irreversibility. That is, the product of the one-step probabilities is different in the two directions along the chain. This is achieved by modeling the transition probabilities in such a way that the local environment is checked for the presence of earlier visited sites more extensively than for the normal self-avoiding walk (S. A. W.).

The first walk (growing S. A. W.–IGSAW) manages to avoid cages and therefore lives forever. We propose to study these walks to describe growth processes of linear polymers in a surrounding where it grows faster than that it can relax to equilibrium (S. A. W.).

We have studied these walks by using different extrapolation methods of enumeration results and extensive M. C. calculations in two and three dimensions. The values of the critical exponent ν and γ of the IGSAW differ considerably from the usual SAW. In two dimensions these values for the GSAW coincide with the SAW values, whereas in three dimensions this is not clear yet.

Paul MEAKIN, *The diffusion-limited growth model: its application to biological growth processes and material breakdown*: The diffusion-limited growth model is a stochastic growth model in which the probability of growth at an unoccupied surface site is a function of the local concentration (C) of some diffusing substance at that site. In the case where the growth itself is a perfect absorber of the diffusing substance which is supplied from a distant source, the diffusion-limited growth model becomes equivalent to the dielectric breakdown model of Niemeyer *et al.*⁽¹⁷⁾ Under these conditions, for the case where the growth probability (P) is related to C by $P \sim C^\alpha$ the model generates structures with fractal dimensionalities of about 1.9, 1.7, and 1.4 for $\alpha \equiv 0.5, 1.0, \text{ and } 2.0$, respectively. Simulations have been carried out with a variety of boundary conditions and growth probability functions $P(C)$ to represent the behavior of biological, chemical, and physical systems.

J. NITTMANN, *A diffusion-limited aggregation model for the viscous finger instability*: The interface between a less viscous fluid displacing a more viscous fluid in a parallel plate flow or in a porous medium is unstable. Instead of a well-defined interface between the two fluids, one observes that the less viscous fluid “tunnels” through the more viscous fluid. A system of long and narrow fingers of the less viscous fluid develops reaching far into the more viscous fluid. Preliminary experimental

investigations indicate that these flow patterns are fractals associated with a Hausdorff dimension. We discuss the application of diffusion-limited aggregation-type models to present the instability patterns.

L. PELITI, *Random walks with memory*: Different models of random walks with memory have been recently introduced in the literature. Some of them may be relevant for the description of polymerization processes. It is possible to give a field theoretical description of these models, leading to definite predictions about their asymptotic behavior. The actual results of simulations cannot however be understood without a proper treatment of the preasymptotic region.

L. PIETRONERO, *Fractal dimension of dielectric breakdown*: It is shown that the simplest nontrivial stochastic model for dielectric breakdown naturally leads to fractal structures for the discharge pattern. The model is based on the Laplace equation associated with a probability field and it gives rise to random fractals with well-defined Hausdorff dimensions. The relations of this model with the diffusion-limited aggregation will be discussed in detail. The possibility of application to other stochastic phenomena like fracture propagation is proposed.

R. RAMMAL, C. TANNOUS, A.-M. S. TREMBLEY, *1/f noise in random resistor networks: fractals and percolating systems*: A general formulation for the spectral noise S_R of linear resistor networks of arbitrary topology is given. General calculational methods, based on Tellegen's theorem are illustrated for one- and two-probe configurations. For self-similar networks, we show the existence of a new exponent b , member of a whole new hierarchy of exponents characterizing the size dependence of the normalized noise spectrum $s_R = S_R/R^2$. b is shown to lie between the fractal dimension \bar{d} and the resistance exponent $-\beta_L$. b has been calculated for a large class of fractal structures: Sierpinski gaskets, X lattices, von Koch structures, etc. For percolating systems s_R is investigated for $p < p_c$ as well as for $p > p_c$. In particular, an anomalous increase of the noise at $p - p_c^+$ is obtained. A finite-size scaling function is proposed and the corresponding exponent b is calculated in mean field theory.

P. RICHETTI, J. PROST, P. BAROIS, *Two-dimensional aggregation and crystallization of a colloidal suspension of latex spheres*: We present experiment investigations of two-dimensional crystallization of calibrated Brownian spheres in water. The interaction monitored by an external AC electric field is attractive in a wide range of frequency and leads to a clustering of fractal crystallites. The fractal dimension of the structure is measured to be $D = 1.76 \pm 0.7$ in good agreement with the theory of dynamical clustering of clusters. Melting occurs upon increasing frequency and is continuous via a phase of hexatic symmetry as seen by light scattering.

L. M. SANDER, *The growing interface of fractal aggregates*: Two lengths can be associated with a growing fractal: λ , the width of the region in which growth occurs, and l , the penetration depth for random walkers incident from the outside. For diffusion-limited aggregation (DLA) they are identical. The exponent by which λ scales with R , i.e., $\lambda \sim R^p$, is discussed. Two hypotheses have appeared: $p = (d - D)/2$ (mean field theory), or $p = 1$ (scaling with a single diverging length). Current numerical evidence, due to Plishke and Racz, shows $p \approx 0.8$ for DLA, and rules out mean field theory. New simulation results are given for DLA in three dimensions in agreement with Plishke and Racz, but with some hints of deviations from the power law.

B. SAPOVAL, M. ROSSO, J. F. GOUYET, *Fractal nature of a diffusion forefront*: Microscopic structure obtained by two-dimensional atomic diffusion from a boundary with constant concentration has been studied by computer simulation. The diffusion frontier defined as the hull of the infinite cluster connected with the source boundary, exhibits a fractal behavior in a range which increases with diffusion depth. Our results accredit the simple-minded idea that the two-dimensional site percolation hull is the limit of the above diffusion frontier for infinite diffusion.

H. Eugène STANLEY, *Fractal concepts in colloids, gels and polymeric materials*: Several new models of aggregation and gelation are described, and are discussed in light of recent fractal concepts.⁽¹⁸⁾ Systematic approaches based on Monte Carlo simulation, exact enumeration, and renormalization group are presented, with emphasis on work by the author and his collaborators.⁽¹⁹⁻³¹⁾

How are the laws of physics modified when the underlying substrate is a fractal? To investigate this question, we first consider the motion of the de Gennes ant, which models electrical transport on a random network (as well as the elastic modulus of a gel). We describe evidence that the two exponents of the problem, d_f (the fractal dimension of the aggregate) and d_w (the fractal dimension of the walk) are possibly related for the ant by the dimension-independent (or “superuniversal”) relation $d_f = (2/3)d_w$, first conjectured for percolation clusters by Alexander and Orbach. We investigate the range of validity of the Alexander–Orbach conjecture and find that it is remarkably large, applying to Witten–Sander aggregates⁽²⁰⁾ and to Toussaint–Wilczek diffusive annihilation⁽²⁶⁾ but *not* to the backbone of a percolation cluster^(24,27,28) or to lattice animals.⁽²⁹⁾ We present an argument, rigorous for the Cayley tree, that supports the Alexander–Orbach conjecture,⁽²¹⁾ and we discuss “how” and “why” that argument may fail by about 2% in $d = 2$.⁽²⁸⁾ A recently proposed model⁽²³⁾ of a correlated walk that may display analogous “superuniversal” behavior of superconducting network (and the viscosity of a gel); here we can relate d_w

to d_f and also obtain a formula for the unscreened perimeter of any fractal.⁽²²⁾ A direct connection is proposed between the “dynamic” transport properties and the “static” topological structure for branched polymers in any spatial dimension d . Specifically, the resistivity exponent ζ is given by $\zeta = d_f/d_1$, where d_1 is the topological dimension (the number of sites within path length l of a given site scales as $M \sim l^d$). To describe the irreversible growth of linear polymers, we introduce a new type of correlated walk, related to the zero initiator concentration limit of the kinetic gelation model.⁽³⁰⁾ We also describe a new geometric model embodying the physical mechanism of the coil-globule transition at the theta point of a linear polymer. We prove an exact mapping between this model and a self-avoiding walk with a monomer-monomer interaction energy $\varepsilon = \ln 2p$, where p is a parameter in the model governing the relative strength of the two-body and three-body interactions.⁽³¹⁾ Finally, recent results on fractal models of polymers are given, including extremely accurate calculations of the fractal dimension of self-avoiding walks, branched polymers, and percolation clusters for $d > 2$.⁽²⁵⁾

H. MARTIN, J. VANNIMENUS and J. P. NADAL, *From invasion to Eden growth: a family of models for cluster growth in a random environment*: We define a family of models to describe cluster growth in random media... These models depend on a temperaturelike parameter and interpolate continuously between the Eden model and the invasion model, recently introduced in the study of flow in porous media. Numerical results are presented in two dimensions, for a version of these models where growth is biased in a given direction. Directed invasion clusters are fractal, with the same exponents as directed percolation clusters, but for the other cases studied the clusters are compact. They remain compact even when a “trapping” rule prevents internal holes from being filled.

D. A. WEITZ, *The aggregation of aqueous colloids: Relationship between dynamics and structure*: The clusters formed by the diffusion-limited, kinetic aggregation of aqueous gold colloids exhibit dilation symmetry. Their fractal dimension is $d = 1.75$, in excellent agreement with the value of 1.8 obtained from computer simulations of the cluster-cluster model. In this talk, we discuss measurements of the dynamics of the aggregation and the evolution of the cluster size distribution. These also compare favorably with the results obtained from computer simulations of the cluster-cluster model. We also discuss experiments aimed at determining the generality of these results. We have varied the nature of the short-range interactions between the individual colloid particles from the purely attractive one, which gives $d_\phi = 1.75$, to one which has a small ($\sim kT$) repulsive barrier that must be overcome before the clusters stick. We find that this causes substantially different aggregation dynamics, qualitatively

different forms of the cluster size distribution, and an apparent but small increase in the fractal dimension. This behavior appears to account for the variation in the experimental values for d_ϕ reported in the literature, and possible approaches to be taken in modeling these results will be discussed. The goal of this work is to determine how the short-range interactions between the particles effect the long-range structure.

T. A. WITTEN, *The role of randomness in diffusive growth*: I discuss the role of random noise in growth models like dendritic crystal growth, viscous fingering, and diffusion-limited aggregation. The time development of all these systems is governed by a field that satisfies Laplace's equation. I report on Yacov Kantor's, Robin Ball's, and my work on a series of models which share this property, but which differ in the role played by random noise. The differences in the spatial scaling properties of these models give information about what features of the noise are relevant to large-scale structure.

ACKNOWLEDGMENTS

We wish to thank C. Moser, director of the CECAM (Centre Européen de Calcul Atomique et Moléculaire), for sponsoring the workshop and for providing us with invaluable help during the course of the meeting.

REFERENCES

1. C. Allain and B. Juhier, *J. Phys. Lett. (Paris)* **44**:L421 (1983).
2. C. Camoin and R. Blanc, *J. Phys. Lett. (Paris)* **46**:L67 (1985).
3. R. Blanc, M. Belzons, C. Camoin, and J. L. Bouillot, *Rheol. Acta* **22**:505 (1983).
4. R. Botet, R. Jullien, and M. Kolb, Hierarchical model for irreversible kinetic cluster formation, *J. Phys. A* **17**:L75 (1984).
5. R. Botet, R. Jullien, and M. Kolb, Gelation in kinetic growth models, *Phys. Rev. A*, **30**:2150 (1984).
6. R. Ball and T. Witten, Particle aggregation versus cluster aggregation in high dimensions, *J. Stat. Phys.* **36**:873 (1984).
7. S. P. Obukhov, Kinetically aggregated clusters, preprint.
8. R. Botet, to appear in *J. Phys. A*.
9. J. B. McLeod, *Q. J. Math. Oxford* **2**:119 (1962).
10. A. A. Lushnikov, *J. Coll. Interf. Sci.* **45**:549 (1973); M. H. Ernst, E. M. Hendriks, and F. Leyvraz, *J. Phys. A: Math. Gen.* **17**:2137 (1984).
11. F. Leyvraz, *Phys. Rev. A* **29**:854 (1984).
12. E. M. Hendriks, M. H. Ernst, and R. M. Ziff, *J. Stat. Phys.* **31**:519 (1983).
13. A. A. Lushnikov and V. N. Piskunov, *Dokl. Akad. Nauk SSSR* **231**:1166 (1976).
14. P. G. J. van Dongen and M. H. Ernst, *J. Stat. Phys.* **37** (1984).
15. R. Jullien and M. Kolb, *J. Phys. A* **17**:L639 (1984).
16. M. Kolb and R. Jullien, *J. Phys. (Paris)* **45**:L977 (1984).

17. L. Niemeyer, L. Pietronero, and H. J. Weismann, *Phys. Rev. Lett.* **52**:1033 (1984).
18. H. E. Stanley, *J. Stat. Phys.* (in press); *Proc. Gaithersberg Fractals Conf., Nov. 1983*: H. E. Stanley and A. Coniglio, *Ann. Israel Phys. Soc.* **5**:121 (1983); H. E. Stanley, F. Family and G. Gould, *J. Poly. Sci.* (in press); H. E. Stanley, *J. Phys. Soc. Japan Supply.* **52**:151 (1983).
19. H. Gould, F. Family, and H. E. Stanley, *Phys. Rev. Lett.* **50**:686 (1983).
20. P. M. Meakin and H. E. Stanley, *Phys. Rev. Lett.* **51**:1457 (1983).
21. F. Leyvraz and H. E. Stanley, *Phys. Rev. Lett.* **51**:2048 (1983).
22. A. Coniglio and H. E. Stanley, *Phys. Rev. Lett.* **52**:1068 (1984).
23. H. E. Stanley, K. Kang, S. Redner, and R. L. Blumberg, *Phys. Rev. Lett.* **51**:1223 (1983).
24. H. E. Stanley and A. Coniglio, *Phys. Rev. B* **29**:522 (1984).
25. I. Majid, Z. Djordjevic, and H. E. Stanley, *Phys. Rev. Lett.* **51**:143 (1983); **51**:1282 (1983); I. Majid and H. E. Stanley, preprint.
26. P. M. Meakin and H. E. Stanley, *J. Phys. A* **17**:L173 (1984).
27. H. J. Herrmann, D. C. Hong, and H. E. Stanley, *J. Phys. A* **17**:L261 (1984).
28. D. C. Hong, S. Havlin, H. J. Herrmann, and H. E. Stanley, *Phys. Rev. Lett.* (submitted)
29. S. Havlin, Z. Djordjevic, I. Majid, H. E. Stanley, and G. H. Weiss, *Phys. Rev. Lett.* (submitted).
30. I. Majid, N. Jan, A. Coniglio, and H. E. Stanley, *Phys. Rev. Lett.* **52**:1257 (1984).
31. A. Coniglio, N. Jan, I. Majid, and H. E. Stanley, preprint.