The Vold–Sutherland and Eden Models of Cluster Formation[1]

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Received March 21, 1983; accepted April 25, 1983

Large clusters or flocs have been grown on the computer using the models introduced by Vold and Sutherland and by Eden. Some of the properties of these clusters have been analyzed and compared with the same properties of clusters grown using the diffusion limited growth process of Witten and Sander. For all three models the radius of gyration (R_g) is related to the number of particles in the cluster (N) by an expression of the form $R_g \sim N^{\beta}$ (in the limit of large cluster sizes). In two-dimensional simulations the Eden (surface growth) model gives compact clusters with a radius of gyration exponent (β) very close to 1/2. For the Vold–Sutherland (linear particle trajectory) model the exponent β has a value close to 1/2 for two-dimensional clusters and close to 1/3 for three-dimensional clusters, if they are sufficiently large. For the Witten–Sander model β is definitely larger than 1/2 in two dimensions (~3/5) and larger than 1/3 in three dimensions (~2/5). Other geometric properties of the clusters have been determined such as the density–density correlation function and the number of particles $N(l)$ within a distance (l) of the center of mass. For the two dimensional Witten–Sander model the dependence of R_g on N, the dependence of $N(l)$ on l, and the density–density correlation function can be described in terms of a single parameter (the Hausdorff dimensionality—D). The significance of the Hausdorff dimensionality is outlined and the concept of Hausdorff dimensionality is used in the discussion of the structures generated using all three models.

INTRODUCTION

The recent work of Witten and Sander on diffusion-limited cluster growth in two-dimensional space (1) and our own extension to higher dimensionalities (2) has stimulated us to examine other models for cluster formation. One of the earliest models for the computer simulation of floc formation in three dimensions was introduced by Vold (3). In this model, particles with random linear trajectories are added to a growing cluster of particles at the position where they first contact the cluster. Reorganization of the cluster is not permitted. One of the most important results of Vold's simulations was that the number of particles within a length l of the center of gravity is given by $N(l) \sim l^{2.33}$, for $N(l) < 40$–60% of the total number of particles in the cluster (N). Vold's work was criticized by Sutherland (4) who pointed out that the procedures used in Vold's simulation did not result in particle trajectories with random direction and position. After correction of this deficiency, Sutherland found that $N(l) \sim l^{2.78}$. Sutherland interpreted this result as "It seems highly probable that as the floc size increases the core reaches a constant porosity of about 0.83." Sutherland also indicated that two-dimensional simulations (with 500 particles per cluster) gave the result $N(l) \sim l^{2.0}$. In this paper, the results of simulations using the Vold–Sutherland (VS) model in two- and three-dimensional space are reported. The clusters used in this study are more than an order of magnitude larger than those simulated by Vold and Sutherland. Our results indicate that $N(l) \sim l^{2.75\pm0.04}$ for three-dimensional Vold–Sutherland clusters and $N(l) \sim l^{1.91\pm0.03}$ for two-dimensional Vold–Sutherland clus-

[1] Contribution No. 3198.

0021-9797/83 $3.00

ters. These results were obtained using clusters of 10,000 particles per cluster and confirm Sutherland's interpretation of his results obtained using much smaller clusters.

We have also investigated the Eden (5) model of cluster formation using a lattice model in which particles are added at random with equal probability to any unoccupied site adjacent to one or more occupied sites. The main emphasis of this paper is the properties of the two-dimensional cluster since it is much easier to approach the limit $N \rightarrow \infty$ in two-dimensional simulations.

The concept of Hausdorff (6) or fractal (7) dimensionality has been used to analyze the results of the computer simulations described in this paper. This idea is a particularly valuable and convenient way of describing many of the geometric properties of structures which are self-similar or statistically self-similar (i.e., structures which have dilation symmetry). In the case of ordinary (compact) objects, it is possible to write down many geometric relationships in terms of the (ordinary Euclidean) dimensionality of the object (d). A few examples are (i) $R_g \sim M^{1/d}$ where R_g is the radius of gyration and M is the mass or volume. (ii) $M(l) \sim l^d$ where $M(l)$ is the mass contained within a distance l of the center of the object. (iii) $P \sim M^{2/d}$ where P is the area of the projection onto a two-dimensional surface.

For more complex structures with a well-defined fractal or Hausdorff dimensionality (D) very similar relationships exist ($R_g \sim M^{1/D}$, $M(l) \sim l^D$, $P \sim M^{2/D} \cdots$ etc.). The Hausdorff dimensionality (D) can be determined from any of these relationships (for example, by measuring the radius of gyration as a function of the mass). Once D has been determined from one of these geometric relationships all the others are known. Another very important quantity which characterizes self-similar objects is the density–density correlation function $C(r)$ (r is distance). If the object has a Hausdorff dimensionality D and an ordinary Euclidean dimensionality d then $C(r) \sim r^{(D-d)}$.

The Witten–Sander model for diffusion-limited aggregation (1) provides a good illustration of how these ideas can be applied. The Hausdorff dimensionality for the statistically self-similar random clusters generated by this model was originally determined from the dependence of the density–density correlation function $C(r)$ on distance (r).

For the case of clusters grown on a two-dimensional lattice ($d = 2$) it was found (1) that $D \approx 5/3$. This result implies that $R_g \sim M^{\beta}$ ($\beta = 1/D = 3/5$) and this result was confirmed by determining the radius of gyration as a function of mass (M) (or the number of particles in the cluster (N)). If $D \approx 5/3$ we also expect that $M(l) \sim l^{5/3}$. In this paper we show that this expectation is valid ($N(l) \sim l^{1.707\pm0.022(\approx 5/3)}$).

One of the main objectives of this paper is to determine if the concept of Hausdorff dimensionality can also be applied to clusters grown using the E and VS models.

It should be noted that in real systems the property of self-similarity or dilation symmetry may extend over only a limited range of length scales. In this paper we are concerned mainly with the structure of clusters on length scales between some lower cut off length and lengths approaching the overall size of the cluster (which may be arbitrarily large). For this reason we are interested in the geometric properties in the limit $N \rightarrow \infty$. If the structure is statistically self-similar, we expect all of the geometric relationships discussed above (and many others) to give the same value for the Hausdorff dimensionality (D). However, if we are not sufficiently close to the $N \rightarrow \infty$ limit, different geometric relationships may give different numerical values for D when they are used to analyze the structure. However, as the $N \rightarrow \infty$ limit is approached, the values for D obtained from different methods of analysis should converge to a single value (D_∞).

The Hausdorff dimensionality has important implications for colloidal systems. If the Hausdorff dimensionality (D) is equal to the Euclidean dimensionality (d) then as the cluster grows larger and larger it will approach a constant limiting density or porosity. However, if $D < d$ (as is known to be the case for the WS model) the density of the cluster will become smaller and smaller as the cluster grows larger and larger. The Hausdorff or fractal dimensionality also has important implications for many other physical properties (8, 9).

The work of Sutherland (4) provides us with an estimate of the Hausdorff dimensionalities of clusters grown using the VS model in two and three dimensions from the dependence of $N(l)$ on l. In two dimensions $N(l) \sim l^{2.0}$ implies $D = d = 2$ and in three dimensions $N(l) \sim l^{2.87}$ implies $D = 2.87$ providing the $N \rightarrow \infty$ limit was approached sufficiently closely.

SIMULATION PROCEDURES

Our general approach to the simulation of floc formation using the mechanism of Vold and Sutherland is to employ the methods of vector analysis (10) (rather than the analytic geometric approach of Vold (2) and Sutherland (3)). The first step is to generate a unit vector with random orientation. This is accomplished by generating three random numbers (two in the case of a two-dimensional simulation) uniformly distributed over the range 0–1. If the vector defined by these three (two) random numbers lies outside of a sphere (circle) of unit radius, it is rejected. Otherwise, the vector is normalized to produce the random unit vector **e**. A second vector **d** with random orientation is generated in the same way, and a vector **b** randomly oriented perpendicular to **e** is obtained from

$$\mathbf{b} = \mathbf{a} \times \mathbf{e}. \quad [1]$$

The vector **b** is normalized to a length of $r_{max} + 1.0$ (where r_{max} is the maximum distance from the center of any particle in the cluster to the origin in units of particle diameters)

$$\mathbf{b}' = \frac{\mathbf{b}}{|b|}(r_{max} + 1.0). \quad [2]$$

Finally another random number (y) is generated ($0 \leqslant y \leqslant 1.0$), and the equation for a random linear trajectory which passes within a distance of $r_{max} + 1.0$ of the origin is given by

$$\mathbf{t} = y^{(1/(d-1))}\mathbf{b}' + s\mathbf{e} \quad [3]$$

or

$$\mathbf{t} = \mathbf{b}'' + s\mathbf{e} \quad (\mathbf{b}'' = y^{(1/(d-1))}\mathbf{b}') \quad [3a]$$

where s is the distance along the trajectory and d is the dimensionality of the space used in the simulation.

Having generated a random trajectory, the next step is to determine which (if any) of the spherical particles of unit diameter have their centers within a distance of 1 particle diameter from **t**. For a point at position **r**, the perpendicular (minimum) distance from **r** to **t** is given by

$$d = (\mathbf{r} - \mathbf{b}'') \times \mathbf{e}. \quad [4]$$

We must now find which of the particles in the clusters whose centers are within a distance of 1.0 from the origin will be first touched by a sphere of unit diameter moving along the trajectory **t**. The position of first contact along the trajectory $\mathbf{t} = \mathbf{b}'' + s\mathbf{e}$ is given by

$$s = \mathbf{r} \cdot \mathbf{e} - (1.0 - d^2)^{1/2}. \quad [5]$$

The simulation is started out with a single spherical particle at the origin, and particles are added to the cluster using the procedure outlined above.

The "Eden" model (5) is so simple that large clusters (100,000 particles per cluster) can be generated with the crudest algorithm. We simply use random numbers to pick lattice sites at random and examine the nearest neighbor sites to determine if a "particle" should be added. The maximum magnitude for the value of any trial coordinate was restricted to $1 + C_{max}$ where C_{max} is the maximum value for the magnitude of any of the coordinates for any lattice site already occupied. To generate even larger clusters using the Eden model, a second algorithm was developed in which a list of unoccupied inter-

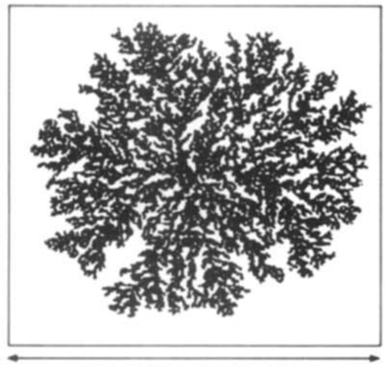

FIG. 1. A typical cluster of 10,000 particles grown in a two-dimensional simulation using the Vold–Sutherland (VS) model.

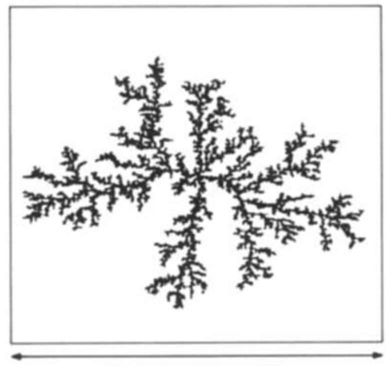

FIG. 3. This figure shows a cluster of 10,000 particles grown on a square lattice using the Witten–Sander model of diffusion limited cluster formation.

face sites is maintained and updated at regular intervals. A site is picked at random from this list and is occupied if it has not been previously occupied since the last update of the list.

RESULTS

Several large clusters (up to 20,000 particles per cluster) were grown in two-dimensional space using the VS model with the procedures outlined in the previous section. Figure 1 shows a typical cluster of 10,000 particles. Using the Eden model, clusters with up to 200,000 particles per cluster were grown. Figure 2 shows a two-dimensional Eden cluster of 10,000 particles. For the purpose of comparison, a two-dimensional cluster of 10,000 particles grown using the Witten–Sander model for diffusion-controlled cluster formation is shown in Fig. 3.

These two-dimensional clusters were analyzed in several ways to obtain estimates of their Hausdorff dimensionalities. For clusters grown by all three mechanisms, the dependence of the radius of gyration (R_g) on the number of particles in the cluster (N) can be expressed as

$$R_g \sim N^{\beta} \qquad [6]$$

for sufficiently large cluster sizes. The Hausdorff dimensionality (D) is given by (11)

$$D = 1/\beta. \qquad [7]$$

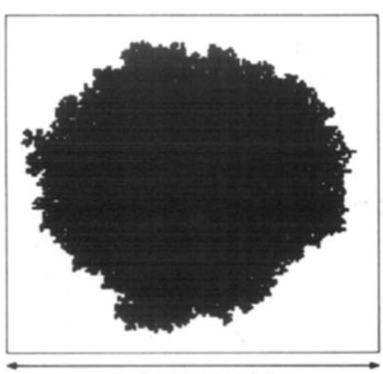

FIG. 2. A cluster of 10,000 particles grown on a two-dimensional lattice using the Eden model.

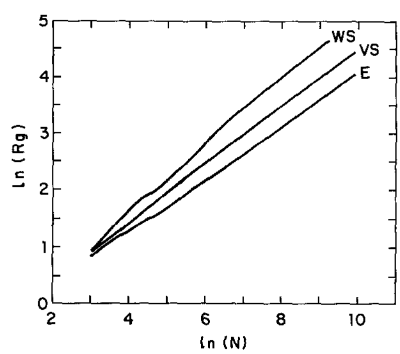

FIG. 4. The dependence of ln (R_g) on ln (N) for the WS, VS, and E models of cluster formation.

TABLE I

Radius of Gyration Exponent (β) Obtained from a Two-Dimensional Vold–Sutherland Model of Cluster Formation

	Cluster size				
	1250	2500	5000	10,000	20,000
	0.487	0.503	0.503	—	—
	0.535	0.512	0.506	0.5086	—
	0.497	0.493	0.509	0.5089	0.5102
	0.501	0.505	0.511	0.5116	—
	0.514	0.519	0.528	—	—
	0.506	0.516	0.507	—	—
	0.527	0.496	0.504	0.5207	0.5137
	0.492	0.523	0.523	0.5122	0.5062
Average	0.507 ± 0.014	0.508 ± 0.009	0.511 ± 0.008	0.512 ± 0.006	0.510 ± 0.0009

Figure 4 shows the dependence of ln (R_g) on ln (N) for the E, VS, and WS models for typical clusters of 10,000 (WS) or 20,000 (VS, E) particles. The radius of gyration exponent β is obtained from a least-squares fit of a straight line to the coordinates (ln (R_g), ln (N)) obtained from the last 50% of intermediate clusters obtained during the formation of a cluster. Results obtained in this manner are given for the VS model in Table I and the E model in Table II. From the clusters of 10,000 particles, a radius of gyration exponent (β) of 0.512 ± 0.006 was obtained for the VS model corresponding to a Hausdorff dimensionality of D (VS) = 1.95 ± 0.002. The results shown in Table II indicate that the *apparent* value for the radius of gyration exponent obtained using the Eden model increases with increasing cluster size. Figure 5 shows a plot of β vs $1/N$. There is no fundamental reason why these data should be plotted in this way. However, if Fig. 5 is accepted at face value, a limiting exponent of $\beta \approx 0.499 \pm 0.001$ ($N \rightarrow \infty$) is

TABLE II

Radius of Gyration Exponents Obtained from the Two-Dimensional Eden Model

N = 100,000–200,000 (6 clusters)	0.4988 ± 0.0004
N = 50,000–100,000 (11 clusters)	0.4984 ± 0.0008
N = 25,000–50,000 (19 clusters)	0.4976 ± 0.0006
N = 12,500–25,000 (26 clusters)	0.4957 ± 0.0008
N = 5,000–10,000 (26 clusters)	0.4925 ± 0.002

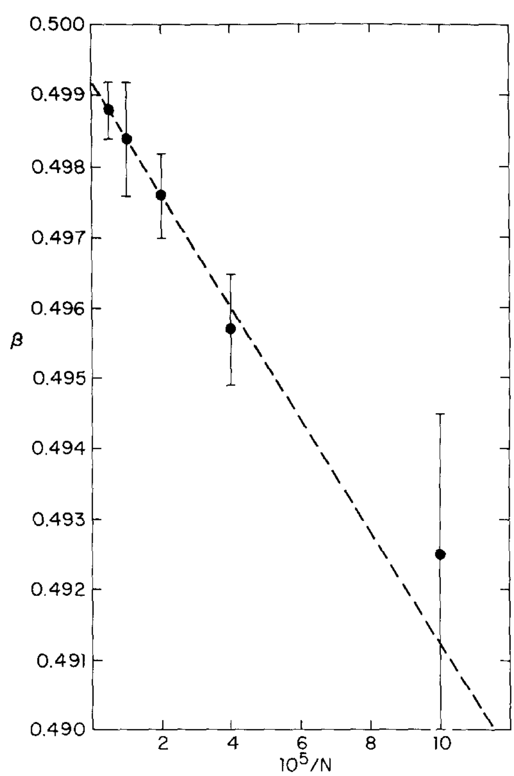

FIG. 5. Dependence of the radius of gyration exponent (β) on cluster size (N) for the Eden model in two dimensions.

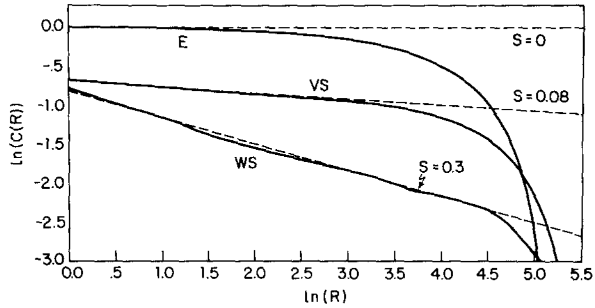

FIG. 6. Typical density-density correlation functions for two-dimensional clusters grown using the Eden (E), Vold–Sutherland (VS), and Witten–Sander (WS) models. The dashed lines indicate the linear relationship between ln ($C(R)$) and ln (R) at intermediate length scales.

obtained. This is quite close to the classical result ($\beta = 0.5$ for a compact cluster).

Another way of estimating the Hausdorff dimensionality is to use the density–density correlation function $C(r)$. The density–density correlation function is given by

$$C(r) = \frac{\int \rho(\mathbf{r}')\rho(r + \mathbf{r}')dr'}{\int \rho(\mathbf{r}')dr'} = N^{-1}\int \rho(\mathbf{r}')\rho(r + \mathbf{r}')d\mathbf{r}' \quad [8]$$

where $\rho(\mathbf{r})$ is the density at position $\mathbf{r}'$ and $\rho(r + \mathbf{r}')$ is the average density at a distance r from $\mathbf{r}'$. For a large cluster with a Hausdorff dimensionality of D, the density–density correlation function for distances r larger than the individual particle size, but considerably smaller than the cluster size, has a power law dependence on r

$$C(r) \sim r^{-\alpha}. \quad [9]$$

The density–density correlation function exponent α is given by $\alpha = d - D$ where d is the Euclidean dimensionality. Typical density–density correlation functions obtained using the VS, E, and WS models are shown in Fig. 6. The results shown in Fig. 6 indicate that the density–density correlation function exponent (α) is much smaller in the E and VS models than in the WS model. For the WS model $\alpha = 0.322 \pm 0.047$ (2). From Fig. 6, we find that α(VS) ~ 0.08 and α(E) ~ 0.0. Since considerably more computer time is required to calculate the density–density correlation function than is required to calculate

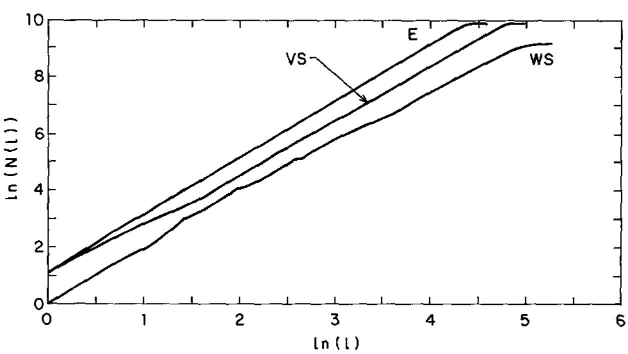

FIG. 7. Dependence of ln ($N(l)$) on ln (l) for three typical clusters grown using the E, VS, and WS models in two dimensions.

TABLE III

Estimates of the Hausdorff Dimensionality (D) for Vold–Sutherland Clusters Grown in a Two-Dimensional Space[a]

	Cluster size			
	2500 ($8.0 \leq l \leq 30.0$)	5000 ($8.0 \leq l \leq 40.0$)	10,000 ($8.0 \leq l \leq 60.0$)	20,000 ($8.0 \leq l \leq 80.0$)
	1.945	1.951	—	—
	1.931	1.939	1.946	—
	1.884	1.887	1.917	1.916
	1.880	1.860	1.876	—
	1.849	1.864	—	—
	1.840	1.874	—	—
	1.864	1.895	1.922	1.924
	1.877	1.877	1.888	1.899
Average	1.88 ± 0.04	1.89 ± 0.03	1.91 ± 0.03	1.91 ± 0.02

[a] D is obtained from a least-squares fit of ln ($N(l)$) vs ln (l).

the radius of gyration as a function of the number of particles in a cluster and some judgment is required in selecting the range of length scales over which the density–density correlation function exponent is determined, we have relied mainly on the radius of gyration to obtain the Hausdorff dimensionality.

A quantity which was determined by Vold (2) and Sutherland (3) is the number of particles $N(l)$ whose centers are within a distance l of the center of gravity of the whole cluster. Figure 7 shows the behavior of ln ($N(l)$) as a function of ln (l) for the VS, E, and WS models. Over intermediate length scales large compared to the size of individual particles and small compared to the size of the cluster, the slope of a plot of ln ($N(l)$) vs ln (l) provides another estimate of the Hausdorff dimensionality (see Introduction). Using nine clusters with an average of 9550 particles per cluster, the Hausdorff dimensionality obtained for the WS model using a two-dimensional square lattice is $D = 1.707 \pm 0.022$ for

TABLE IV

Radius of Gyration Exponents (β) Obtained during the Formation of Vold–Sutherland Clusters in Three-Dimensional Space[a]

	Cluster size				
	1250	2500	5000	10,000	20,000
	0.3455	0.3175	0.3408	—	—
	0.2921	0.3387	0.3355	0.3360	—
	0.3276	0.3129	0.3331	0.3369	0.3401
	0.3258	0.3436	0.3416	0.3460	—
	0.3191	0.3537	0.3544	—	—
	0.3040	0.3211	0.3420	0.3410	0.3353
	0.3566	0.3448	0.3274	0.3266	0.3373
Average	0.324 ± 0.020	0.333 ± 0.015	0.339 ± 0.008	0.337 ± 0.009	0.338 ± 0.006

[a] The last 50% of the intermediate clusters are used to calculate the radius of gyration exponents.

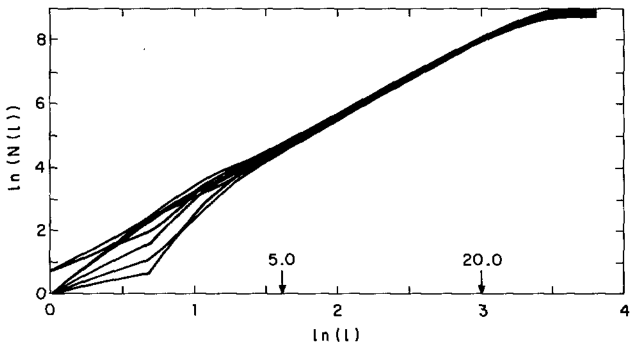

FIG. 8. Dependence of ln ($N(l)$) on ln (l) for eight clusters grown using the WS model of diffusion-limited cluster formation on a three-dimensional cubic lattice.

$10.0 < R < 100.0$. This result is in good agreement with our earlier results obtained from the radius of gyration (2) (1.73 ± 0.06) and from the density–density correlation function (1.68 ± 0.05). For the Eden model, the center of the cluster is compact and the dependence of ln ($N(l)$) on ln (l) gives a Hausdorff dimensionality of exactly 2.0. The results obtained using the VS model are shown in Table IV. The results shown in Table III indicate that $D = 1.90 \pm 0.03$ for the VS model.

Similar simulations have been carried out in three-dimensional space using the VS and WS models. We did not carry out simulations using the Eden model since we expect that this model will give a compact cluster ($d = D = 3.0$) as is the case in the two-dimensional simulations: From nine clusters (average 7600 particles per cluster) a radius of gyration exponent (β) of 0.397 ± 0.012 was obtained using the Witten–Sander model. This result is in good agreement with our earlier simulations ($\beta = 0.402 \pm 0.009$) (2). The corresponding Hausdorff dimensionality obtained by this method is $D = 1/\beta = 2.52 \pm 0.08$. The Hausdorff dimensionality has also been obtained from the dependence of $N(l)$ on l. The results obtained for eight of the clusters are shown in Fig. 8. For $5.0 \leqslant 1 \leqslant 20.0$, an estimate of the Hausdorff dimensionality ($D = 2.43 \pm 0.04$) is obtained. For $5.0 \leqslant 1 \leqslant 15$, we find $D = 2.46 \pm 0.04$.

Several large clusters (up to 20,000 particles per cluster) were grown using the VS model in three-dimensional space. A typical cluster of 5000 particles is shown in Fig. 9. The Hausdorff dimensionality associated with these clusters has been obtained from both the dependence of the radius of gyration on cluster size and the number of particles $N(l)$ whose centers are within a distance l of the center of mass. Values for the radius of gyration exponents (β) obtained from the last 50% of the intermediate clusters generated during the production of our clusters are shown in Table IV. Estimates of the Hausdorff dimensionality obtained from the dependence of ln ($N(l)$) on ln (l) are given in Table V.

DISCUSSION

In this paper, we compare the properties of clusters grown in two and three dimen-

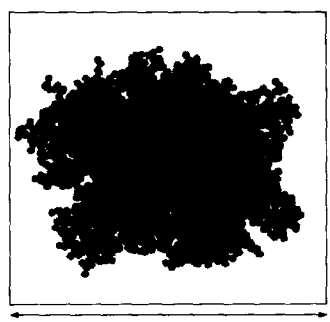

FIG. 9. A cluster of 5000 particles grown in three dimensions using the Vold–Sutherland model.

TABLE V

Estimates of the Hausdorff Dimensionality (D) for Vold–Sutherland Clusters Grown in a Three-Dimensional Space Using the Dependence of ln ($N(l)$) on ln (l)

	Cluster size			
	2500 ($3.0 \leq l \leq 9.0$)	5000 ($3.0 \leq l \leq 12$)	10,000 ($5.0 \leq l \leq 15.0$)	20,000 ($5.0 \leq l \leq 20$)
	2.619	2.705	—	—
	2.848	2.862	2.764	—
	2.600	2.649	2.772	—
	2.740	2.754	2.730	2.788
	2.745	2.743	—	—
	2.667	2.737	2.799	2.806
	2.605	2.655	2.709	2.737
Average	2.69 ± 0.09	2.73 ± 0.07	2.75 ± 0.04	2.78 ± 0.09

sions using the Eden, Vold–Sutherland, and Witten–Sander models. Eden clusters grown on two-dimensional lattices rapidly develop a central region which becomes completely dense and occupies a larger and larger fraction of the total cluster as the cluster grows. This observation indicates that for this model we have $D = d = 2$. This conclusion is confirmed by our numerical studies which indicate that $D \sim 2.003 \pm 0.003$ from the dependence of the radius of gyration on the number of particles in the cluster, $D \sim 2.0$ from the density–density correlation function, and $D = 2.0$ (almost exactly) from the dependence of $N(l)$ on l. In two-dimensional simulations, we find that the Vold–Sutherland model gives a Hausdorff dimensionality of about 1.95 from the dependence of ln (R_g) on ln (N) and 1.90–1.95 from the density-density correlation function. The dependence of ln ($N(l)$) on ln (l) gives $D \sim 1.90$. All of these results taken together indicate that $d - D$ is small but finite, i.e., $d - D \sim 0.05$–0.1. It should be noted that our results have at most a very small dependence on cluster size over the range 2500–20,000 particles per cluster. However, we cannot exclude the possibility that *very* much larger clusters would give estimates for the Hausdorff dimensionality closer to the Euclidean dimensionality. It is clear that this question will not be easily resolved by further computer simulations.

In three dimensions, we find $D \sim 2.97 \pm 0.08$ for the VS model using the dependence of the radius of gyration on the number of particles in the cluster. From the dependence of $N(l)$ on (l), a value of 2.75 ± 0.04 is obtained for clusters of 10,000 particles. In this case, the radius of gyration gives results which seem to be almost independent of cluster size (N) over the range $2500 < N < 25{,}000$. Our estimates for D obtained from the dependence of $N(l)$ on l increase (slowly) with increasing cluster size. Taken together, these results indicate that in the limit $N \rightarrow \infty$ a value close to 3.0 would probably be obtained. Consequently, in both two and three dimensions, we conclude that the VS model gives clusters with a Hausdorff dimensionality close to the Euclidean dimensionality and that the possibility that $D = d$ ($d = 2, 3$) cannot be excluded.

In contrast, the Witten–Sander model gives estimates for the Hausdorff dimensionality which are clearly smaller than the Euclidean dimensionality ($D \sim 1.70$ for $d = 2$ and $D \sim 2.50$ for $d = 3$). It should also be noted that the estimates for D obtained using different methods differ by amounts which are considerably larger than their associated statistical uncertainties. This is not surprising, but it does illustrate the uncertainties associated with the use of numerical simulations to obtain the Hausdorff dimensionality of structures such as the clusters simulated in this paper. It is probable that different methods for obtaining the Hausdorff dimensionality approach the $N \rightarrow \infty$ limit in different ways as the cluster sizes increase.

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